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AN APPARATUS FOR AUTOMATICALLY PLOTTING ELECTRON TRAJECTORIES

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The electrolytic tank is a versatile tool for determining the paths of electrons in an electric field (e.g. in an electron tube or an electrostatic lens). Generally, the procedure is to use the tank to determine the equipotential surfaces, from which the electron trajectories are then construed step by step. The apparatus described here, which is based on a principle put forward by Gabor and Langmuir, traces out the trajectories automatically. A trolley, riding on a drawing board over the tank, is controlled by the voltages from a number of probes (in this case four) in such a way as to cause a stylus to plot the trajectory on the board.

When designing the electrode system of an electrostatic lens, for example, or an electron gun for a cathode-ray tube, it is important to be able to predict the motion of the electrons in the system.

For this purpose one can determine the potential distribution in the system (e.g. with an electrolytic tank or a resistance network) and from this construe the electron trajectories. This method, however, is very cumbersome. The apparatus described in this article traces out the electron trajectories automatically. The field strength and the voltage, quantities that are needed for calculating the trajectories, are measured with an electrolytic tank. A trolley to which a stylus is attached rides on a drawing board above the tank. From the measured quantities at each position the apparatus computes the curvature which the track of the trolley must have at that position in order for the stylus to trace out the trajectory of an electron.

The electrolytic tank

We shall first briefly describe the electrolytic tank as used for determining the potential distribution in an electrode system¹⁾. The tank is filled with a weakly conducting liquid — the electrolyte (usually water) — in which a model of the electrode system is placed. When voltages are applied to the electrodes of the model, a potential distribution is produced in the electrolyte which is independent of

the nature of the dielectric, and thus identical with the distribution that would be found in a vacuum.

Use is now made of a similarity rule which states that the shape and relative potential of the equipotential surfaces do not change when the voltages between the electrodes are multiplied by an arbitrary factor, or when the electrode system is made proportionally larger or smaller. Making use of this, it is possible to work with conveniently low voltages, lower by a suitable factor than in the actual system. The factor need not remain constant, that is to say it is possible to use alternating voltage. This is in fact done in most cases, with the object of avoiding polarization at the electrodes. The dimensions of the electrodes are generally chosen larger than in the actual system, which improves the accuracy of the plot.

For practical reasons, the electrolytic tank is used almost exclusively for measurements in symmetry planes of an electrode system (see the first of the two articles mentioned under reference¹⁾). To this end the model is mounted in the tank in such

¹⁾ See e.g. K. F. Sander and J. G. Yates, The accurate mapping of electric fields in an electrolytic tank, *Proc. Instn. Electr. Engrs.* **100 II**, 167-175, 1953. See also: G. Hepp, Measurements of potential by means of the electrolytic tank, *Philips tech. Rev.* **4**, 223-230, 1939; N. Warmoltz, Potential distribution at the igniter of a relay valve with mercury cathode, *Philips tech. Rev.* **8**, 346-352, 1946; F. Reiniger, The study of thermal conductivity problems by means of the electrolytic tank, *Philips tech. Rev.* **18**, 52-60, 1956/57.

a way that its plane of symmetry coincides with the surface of the electrolyte (fig. 1). The potentials in this plane are measured with a pin probe dipped in the electrolyte.

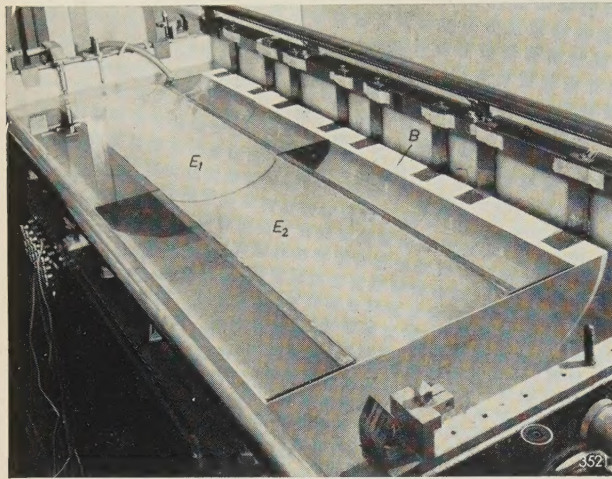


Fig. 1. The electrolytic tank containing a model of an electrostatic lens consisting of two electrodes. The system possesses rotational symmetry; the measurement is made in a plane through the axis of the system. The model must be mounted in such a way that this plane coincides with the surface of the liquid. For this purpose the electrodes E_1 and E_2 rest in a semi-cylindrical plastic trough B , which is suspended from the edge of the tank and can be adjusted vertically by means of screws.

Principle of the automatic plotter

When a particle of mass m and charge $-e$ is accelerated from rest by a potential difference V to a velocity v , its kinetic energy is given by $\frac{1}{2}mv^2 = eV$. The curvature of the trajectory is determined at every point by the instantaneous velocity of the particle and the force F_n acting upon it in a direction perpendicular to the trajectory. The radius of curvature ϱ is:

$$\varrho = \frac{mv^2}{F_n} = \frac{2eV}{eE_n} = \frac{2V}{E_n}, \quad \dots (1)$$

where V is the potential difference between the point of interest and the place where the velocity of the particle was zero, and E_n is the field-strength component perpendicular to the trajectory at the position of the particle.

The trajectory is plotted with the aid of a three-wheel trolley which rides on a drawing board above the tank. A sketch of the trolley is given in fig. 2. The stylus is located under point O on the rear axle. The trolley is propelled by an electric motor which drives the front wheel. The angular position of the front wheel is changed by turning the steering shaft St (perpendicular to the plane of the drawing). If the distance from the steering shaft to the rear

axle is h , and the angle between front and rear axles is α , the stylus describes a circle whose radius is

$$\varrho = \frac{h}{\tan \alpha} \dots \dots \dots (2)$$

The stylus thus traces the trajectory of the particle, provided the following condition is satisfied:

$$\frac{h}{\tan \alpha} = \frac{2V}{E_n},$$

or, otherwise expressed,

$$\alpha = \tan^{-1} \left(\frac{hE_n}{2V} \right) \dots \dots \dots (3)$$

At any given point, then, α must be adjusted in accordance with the values of E_n and V at that point.

This principle was independently described by D. Gabor ²⁾ and D. B. Langmuir ³⁾ as early as 1937. They determined the potential difference V and the field strength E_n from the potentials of two probes. In our case, use is made of four probes in line, which results in greater accuracy, the curve which the potential variation describes along the line through the probes now being approximated by a third-degree function instead of by a linear function. The probes are mechanically linked with the trolley in such a way that they always remain vertically below the rear axle. They are mounted symmetrically with respect to the stylus at equal distances apart (see fig. 2). Voltages proportional to V and E_n are applied to a servo system driving a servo motor

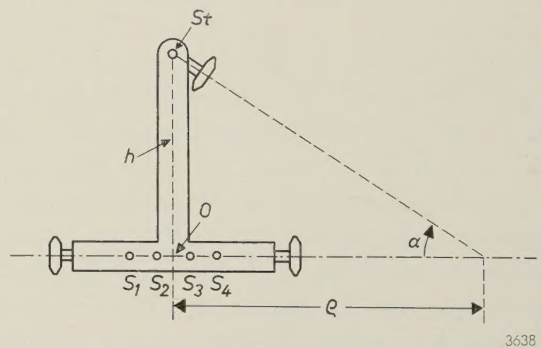


Fig. 2. Sketch of the three-wheel trolley which tracks the electron trajectories. The stylus is located under point O of the rear axle. $S_1 \dots S_4$ are the positions where, under the board on which the trolley rides, four probes dip into the electrolyte. The front wheel is driven by an electric motor. The steering shaft St (vertical shaft which turns the front axle) is controlled by a servo motor so as to cause the stylus to trace out the trajectory.

²⁾ D. Gabor, Mechanical tracer for electron trajectories, Nature **139**, 373, 1937.
³⁾ D. B. Langmuir, Automatic plotting of electron trajectories, Nature **139**, 1066-1067, 1937, and An automatic plotter for electron trajectories, R.C.A. Rev. **11**, 143-154, 1950.

which controls the angular position of the steering shaft of the trolley.

It is possible to start plotting a trajectory at any arbitrary point. The stylus of the trolley is then simply placed above that point. The direction of the initial velocity at which the electron travels at the starting point can also be arbitrarily chosen by facing the trolley in a particular direction. (The tangent of the trajectory coincides with the line h in fig. 2, which thus gives the direction in which the electron is moving.) The speed of the trolley — which is of course in no way related to the velocity of the electron — may be freely chosen; the trolley can also be stopped and reversed for the purpose of checking the trajectory described.

General description of the apparatus

A schematic diagram of the complete equipment is shown in fig. 3. An RC generator delivers a sinus-

applying to up to four other electrodes of the model. The voltages of the four probes S are supplied via cathode followers KV to two computing circuits. Circuit VR determines from these four voltages the potential V midway between the probes (i.e. at the position of the stylus). It further delivers the voltage $-V$, which is also required for the servo system. Circuit ER determines from the four probe voltages the field strength E_n at the position of the stylus.

The servo system consists of a potentiometer circuit, an amplifier and a two-phase servo motor. The potentiometer circuit is mounted on the trolley W , and consists of two potentiometers and some fixed resistors. This circuit is supplied with the voltages V , $-V$ and gE_n (g is a constant having the dimensions of length, and thus gE_n has the dimensions of voltage; the method of calculating g is discussed below). The wipers of the potentiometers are mechanically coupled to the steering shaft on

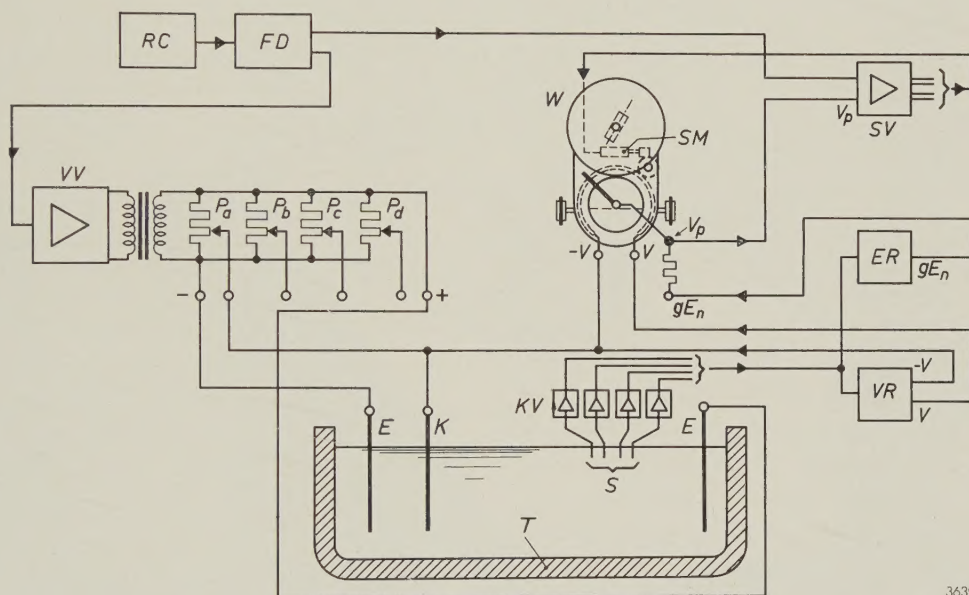


Fig. 3. Schematic diagram of the automatic plotter. RC generator for 500 c/s. FD phase shifter. VV power amplifier. $P_a \dots P_d$ potentiometers for adjusting the supply voltages for the electrodes E . T electrolytic tank. K cathode of electrode system, i.e. the electrode at which the electrons have zero velocity. S probes. KV cathode followers. VR computing circuit for determining the average probe potential V . ER computing circuit for determining the local field strength E_n . W trolley with potentiometer circuit and servo motor SM . SV servo amplifier.

oidal alternating voltage of about 500 c/s. This is supplied via a phase shifter FD , whose function will presently be explained, to a power amplifier VV , which delivers a square-wave voltage to an output transformer, the secondary of which is floating. Connected to this secondary are four potentiometers, so that from the total voltage, which is applied across two electrodes of the model, four variable intermediate voltages are available for

the trolley. The circuit is so designed as to give no output signal when the steering shaft is correctly aligned. If there is a deviation, the potentiometer circuit delivers a voltage V_p , which, via the amplifier SV , actuates the servo motor SM . For the sake of accuracy it is necessary (see later) to use the voltage V_p only during a small part of each cycle. The phase of this part is adjusted with the aid of the second output voltage from the phase shifter FD .

Fig. 4 shows a general view of the apparatus, in the process of plotting a trajectory in an electron gun for a television picture tube. The electronic equipment is not visible in this photograph.

Tank and electrodes

The electrolytic tank is made of reinforced con-

crete, 192×78 cm and 50 cm deep. The walls are clad with a layer of polyethylene 3 mm thick, to prevent ions from the concrete dissolving in the water.

The surface of the probes and electrodes are subject to effects which may adversely influence the accuracy. When a metal is brought into contact with

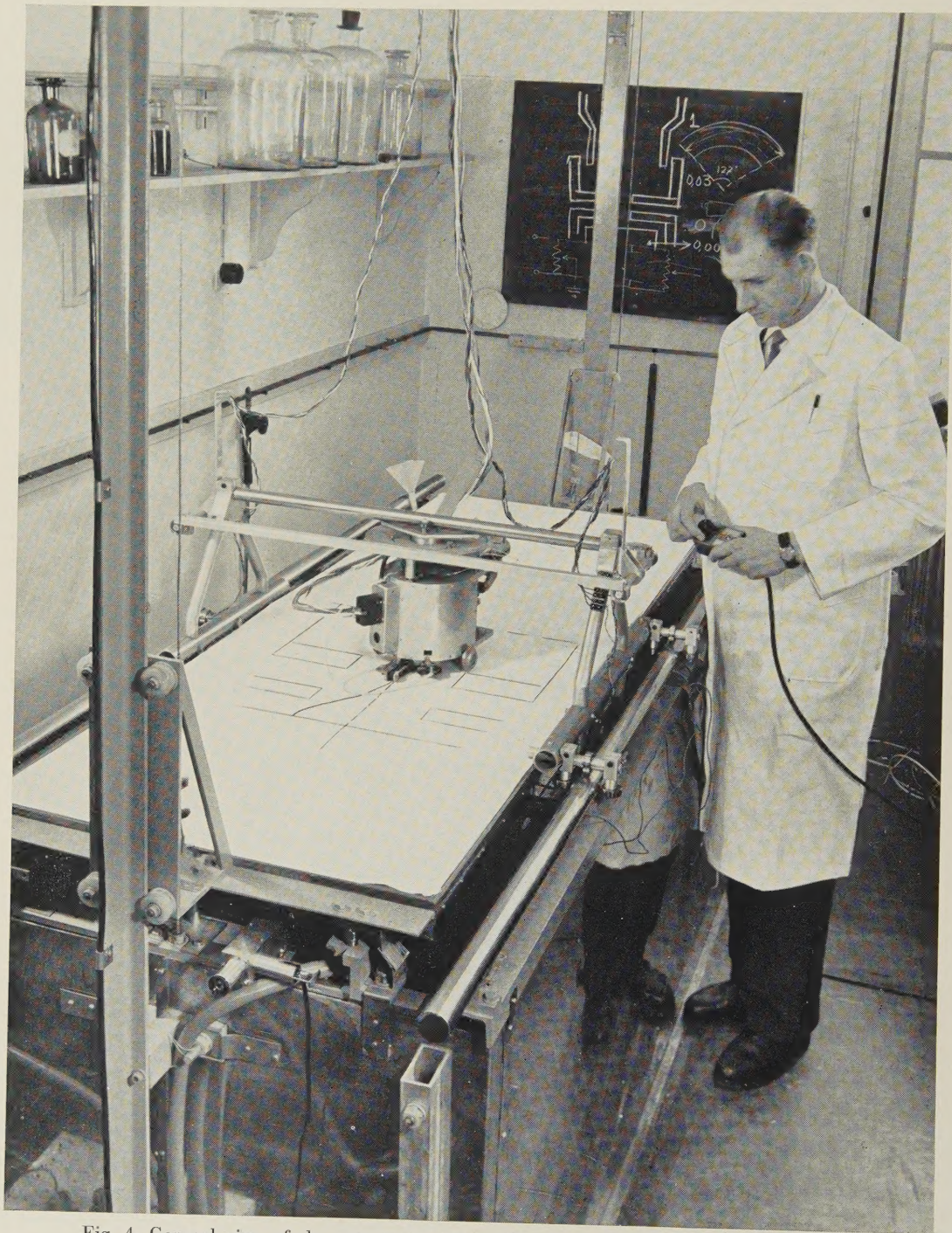


Fig. 4. General view of the automatic plotter. The control box held by the laboratory assistant contains a switch for reversing the direction of the trolley, and a potentiometer for controlling the speed of the trolley. A trajectory is here being plotted in an electron gun for a television picture tube (to make the trace visible in the photograph the trajectory plotted has been traced over by hand). The form of the electrodes is marked out on the drawing board. When all the trajectories have been plotted, the board is photographed and then cleaned. To enable the model to be set up in the tank the whole board and appendages are hoisted up the vertical beams.

an electrolyte, a potential difference arises between the metal and the electrolyte, and if a current flows between them an additional potential difference, due to polarization, may arise. This effect depends on the current density. The polarization of the probes and electrodes must be kept to a minimum; the constant potential difference then remaining is harmless, owing to the use of alternating voltage. Polarization can be reduced to negligible proportions by covering the metal surface with platinum black, i.e. a porous layer of very finely divided platinum. This is in fact done in the case of the probes, but the electrodes are too large to make such a coating practicable. In their case, silver plating was found to result in a sufficiently low polarization, and the electrodes are therefore made of silver-plated brass or copper.

The effect of polarization on the potential measurements can be further reduced by using an electrolyte of high resistivity. In our case, use is made of deionized water, whose resistivity is roughly 30 k Ω cm as against the 2 k Ω cm of main water.

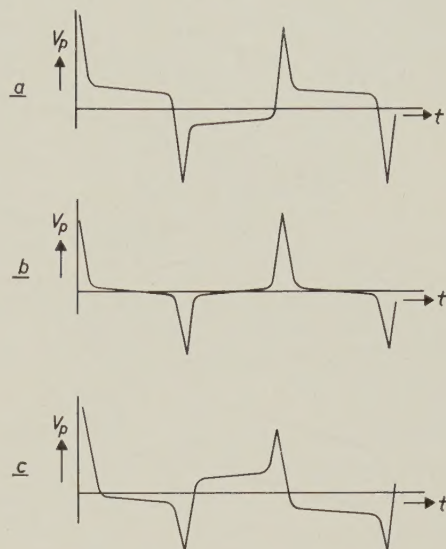
To achieve maximum accuracy, two further conditions must be satisfied.

- 1) The electrolyte must be uniformly conductive. This can be assured simply by stirring the electrolyte shortly before the measurement.
- 2) The probes must not draw current from the electrolyte, as this would disturb the potential distribution. For this reason the probes are connected to cathode followers having an extremely high input resistance.

As a result of polarization of the electrodes, a phase shift occurs between the voltage on the electrodes and the current; consequently, when a sinusoidal voltage is used there is no clearly defined zero point in the output voltage V_p of the potentiometer circuit. V_p is a linear combination of gE_n and V . The voltage gE_n is calculated from the potential difference between the probes, that is between points in the electrolyte, whereas V is the potential difference between a point in the electrolyte and an electrode of the model (the cathode of the system). The phase shift is therefore not felt in gE_n , but it does affect V . These two voltages do not therefore have the same phase, and so they can never be zero at the same time and V_p can never be zero.

As a method of obtaining a sharp zero-point setting, Sander and Yates⁴⁾ have proposed the use of a square-wave voltage for the electrodes of an electrolytic tank. This is done in the present equipment; the voltage V_p then has the waveform shown

in fig. 5a, b and c, depending on the position of the wiper of the potentiometer on the trolley. It is now possible to make a sharp adjustment to the equilibrium setting (fig. 5b) by using a circuit that responds only to the flat portions (plateaus) of the waveform.



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Fig. 5. Output voltage V_p of the potentiometer circuit when the electrodes are supplied with a square-wave voltage.

b) The steering shaft of the trolley is in the correct position. The middle of the "flat" portion (plateau) of the curve is zero. a) and c) The steering shaft is not in the correct position. The voltage of the middle of the plateau is amplified and fed to the servo motor, which corrects the position of the steering shaft.

Excitation of the field in the tank

As we have seen, the electrodes are fed with a square-wave alternating voltage, whilst in the actual system the electrodes are at positive or negative DC potentials. When the electrodes are connected to the output of the power amplifier VV (fig. 3), one of the output terminals is regarded as positive and the other as negative. Interchanging these terminals would imply that the phase of the voltages on the electrodes (and therefore also the phases of V , E_n and V_p) would be shifted 180°. It will be shown when dealing with the servo amplifier that this would make the servo system unstable.

As an example of adjusting the voltages on the electrodes, we shall consider a triode where $V_c = 0$ V, $V_g = -10$ V, and $V_a = +200$ V. The grid is connected to the "negative" terminal of the power amplifier, and the anode to the "positive" terminal. The cathode must now be given a potential, relative to the negative terminal, amounting to 10/210 times the output voltage of the amplifier. For this purpose the cathode is connected to the wiper of potentiometer P_a in fig. 3. For pre-setting the wiper to obtain the required potential, a bridge circuit

⁴⁾ See the first article quoted under reference 1).

is used as shown in *fig. 6*. An accurately calibrated decade potentiometer P_{dec} is connected, in parallel with P_a , to the amplifier. The wiper of P_{dec} is adjusted until the resistances of the parts into which

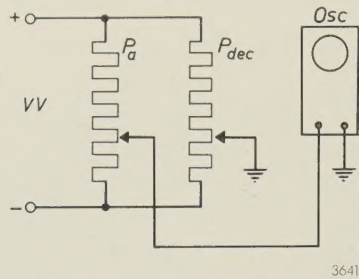


Fig. 6. Bridge circuit for adjusting the potentiometers from which the electrode potentials are tapped. VV output of power amplifier. P_a potentiometer to which the electrode will be connected. P_{dec} calibrated decade potentiometer, pre-set to the required voltage ratio. *Osc* oscilloscope.

the potentiometer is divided are in the ratio of 10 : 210. The wiper of P_{dec} is earthed, and that of P_a is adjusted with the aid of an oscilloscope until its potential is also zero.

For reasons which will be discussed in connection

with the computing circuit for E_n , the potential midway between the probes must be zero. This is achieved by deducting the voltage V from all electrode voltages, the method being to connect the output $-V$ of the V -computing circuit to the electrode of the system for which the electron velocity is zero, as illustrated in *fig. 3*. This electrode is referred to as the cathode, irrespective of its function in the actual system. If the system contains no such electrode, one of the potentiometer terminals must nevertheless be set at zero potential and connected to the output $-V$. Take, for example, an electron lens system with two electrodes where $V_2 > V_1 > 0$. The zero voltage is taken from the negative terminal, and electrode 2 is connected to the positive terminal. Electrode 1 is connected to the wiper of P_a , which is adjusted to the ratio V_1/V_2 .

Method of determining E_n and V

The field-strength component E_n and the potential V may in principle be determined, as Gabor and Langmuir have done, from the voltages of two probes at either side of the measuring point, along

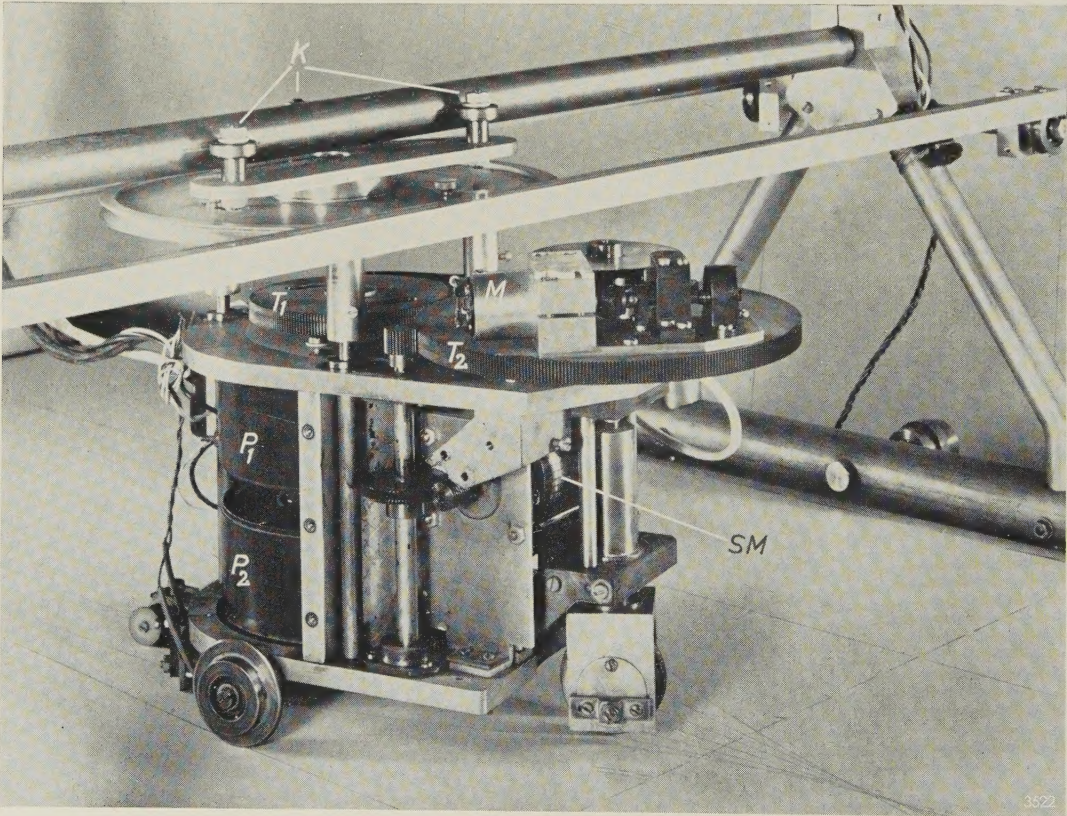


Fig. 7. The trolley on the drawing board. M motor which drives the front wheel for propelling the trolley. SM servo motor which controls the angular position of the steering shaft via a worm and pinion and gear T_2 . P_1 and P_2 potentiometers of servo system. T_1 and T_2 gear wheels which mechanically couple the potentiometer wiper arms to the steering shaft. K ball bearings which transmit the length-wise movement of the trolley to a cross-frame that carries along the probes under the drawing board. (The cover-plates protecting the potentiometers in *fig. 4* have here been removed.)

a line perpendicular to the direction of the trajectory. If the distance between the probes is a and their voltages are V_1 and V_2 , then $E_n = (V_1 - V_2)/a$ and $V = \frac{1}{2}(V_1 + V_2)$. The voltages might be determined by means of a difference amplifier and a sum amplifier. However, at points where the field strength depends markedly on position, the values found in this way would not be accurate. The error can be reduced, of course, by reducing the distance between the probes, but in that case the difference between the voltages is smaller, and therefore higher demands must be made on the accuracy with which the voltages are measured. Moreover, the local disturbance which the probes cause in the potential distribution is then greater.

As we have said, the instrument here described uses four probes, making it possible to determine E_n and V with greater accuracy without reducing the distance between the probes. Of course, the formulae defining E_n and V are more complicated, and more elaborate electronic equipment is needed for determining these voltages.

Before dealing at length with the computing circuits and the derivation of the formulae, we shall touch briefly on the components of the servo system.

The trolley

A photograph of the three-wheel trolley with which the electron trajectories are plotted is shown in *fig. 7*. The trolley is propelled by an electric motor M which drives the front wheel. A servo motor SM turns, via a wormwheel and pinion, a gearwheel T_2

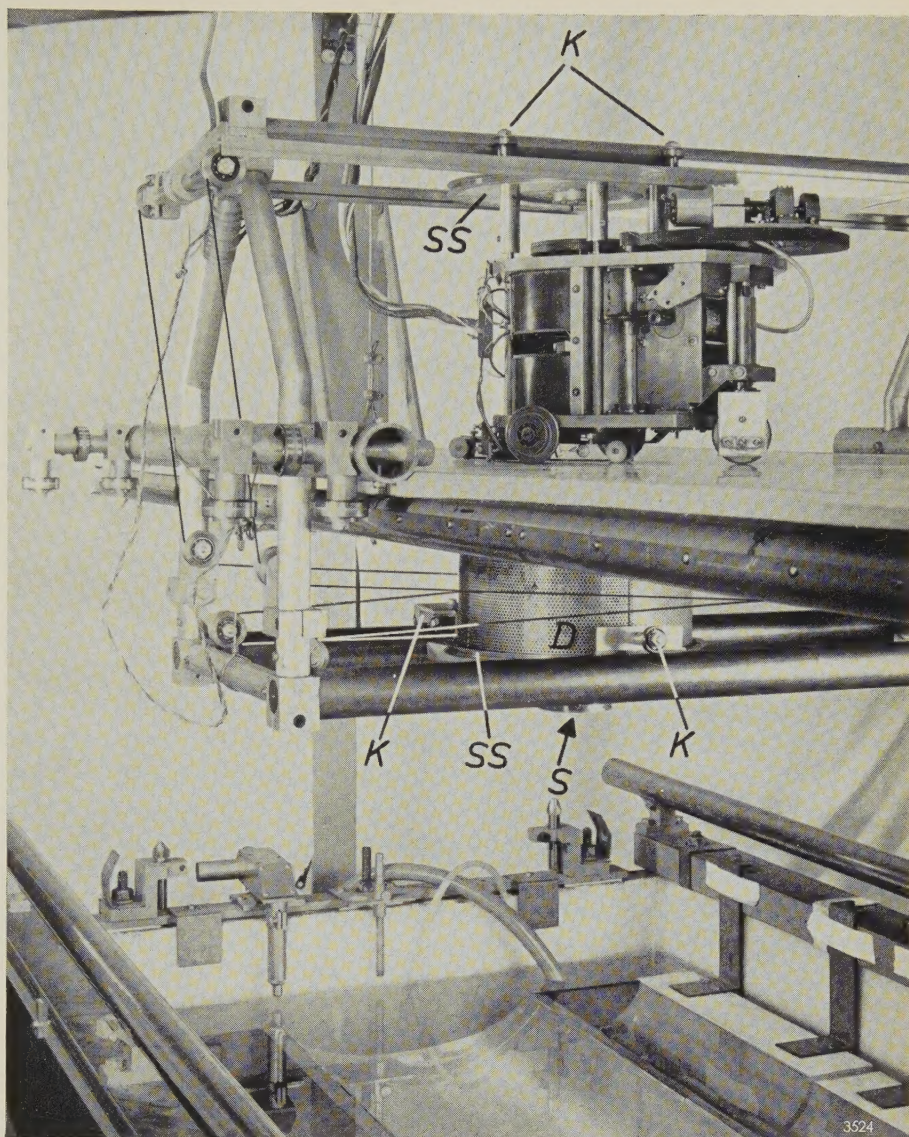


Fig. 8. In this photograph the drawing board has been raised to show the cross-frame and components underneath the drawing board. The box D can be seen, which contains the four cathode followers, and under it the probe holder with the four probes S . These protrude underneath, but are too small (0.5 mm diameter) to be visible.

The movement of the trolley along the length of the tank is transmitted by ball bearings K to the cross-frame, which, again by ball bearings, transmits this movement to the box D . The lateral and rotational movements of the trolley are transmitted by cords passing over the large pulleys SS . Underneath the trolley can be seen the tracing mechanism, consisting of a relay, two rollers over which a typewriter ribbon runs and the wheel which draws the line.

fixed to the steering shaft. Mounted on the trolley are the two potentiometers, P_1 and P_2 , of the potentiometer circuit. With the aid of gearwheels T_1 and T_2 the two wiper arms are mechanically coupled to the steering shaft. In *fig. 8* the drawing board has been raised to reveal various parts situated under it. Here are located the probes S in the probe-holder, which is mounted on a box D containing the four cathode followers. The probes must at all times follow the trolley in both position and orientation; all the movements of the trolley must therefore be transmitted to them. Along the main axis of

the tank this is done by means of a cross-frame which travels along the rim of the tank, and against which the trolley and the box abut via ball bearings *K*. The trolley further carries a large pulley *SS*, over which two cords run via small pulleys to an identical large pulley fixed to the box underneath the drawing board. This mechanism, then, is responsible for transmitting the lateral and rotary movements of the trolley to the probes.

The recording mechanism is visible under the trolley in fig. 8. It comprises an endless typewriter ribbon which passes over two rollers. A fairly sharp-rimmed wheel, attached to the armature of a relay, presses the ribbon on to the drawing board when this relay is energized. Being stationary relative to the drawing board, the ribbon rolls continuously as the trolley moves.

Potentiometer circuit

The principle of the potentiometer circuit mounted on the trolley is illustrated in fig. 9*a*. The deviation of the wiper of potentiometer *P*₁ from the centre point, denoted *x*, may vary from $-\frac{1}{2}$ to $+\frac{1}{2}$. Since *a* (the angle between the front and rear axles of the trolley) must be able to run from $-\frac{1}{2}\pi$ to $+\frac{1}{2}\pi$, the transmission between the potentiometer spindle and the steering shaft must be chosen such that $a = \pi x$.

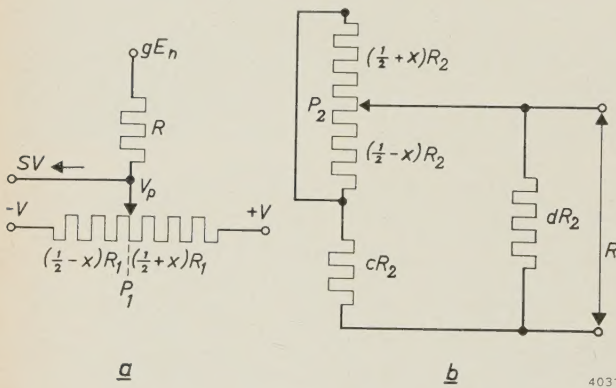


Fig. 9. *a*) Principle of the steering circuit mounted on the trolley. The potentiometer *P*₁ is supplied with the voltages *V* and $-\bar{V}$, and the resistor *R* with the voltage *gE*_n. The position of the potentiometer wiper arm is determined by the angular position of the steering shaft. The voltage *V*_p of the wiper is applied to the servo amplifier *SV*.
b) Circuit which ensures that *R* depends according to a certain function on the position of the steering shaft, whereby *V*_p is zero when the steering shaft is in the correct position. The wiper arm of potentiometer *P*₂ is coupled mechanically to that of potentiometer *P*₁ in (*a*).

When the wiper of the potentiometer is in either of its extreme positions, a small resistance still remains between the wiper contact and the connection terminal, so that *x* cannot become precisely $\frac{1}{2}$ (or $-\frac{1}{2}$). In the extreme positions, then, the angle *a* remains somewhat smaller than $\frac{1}{2}\pi$, and the radius

of curvature of the trajectory cannot therefore be zero. The minimum radius of curvature that can be described by the trolley is 3 mm. For most practical purposes this is quite sufficient. If an even stronger curvature is required, part of the model can be made on a larger scale and the trajectory can then be described in that particular part.

From Kirchhoff's first law, $\Sigma i = 0$, it follows that:

$$V_p = \frac{(\frac{1}{4} - x^2)R_1 g E_n - 2x R V}{(\frac{1}{4} - x^2)R_1 + R} \quad \dots (4)$$

This voltage is used as the output voltage of the circuit and must therefore be zero when the steering shaft is in the correct angular position. In that position, $a = \tan^{-1} (hE_n/2V)$, and therefore *V*_p must be zero when $\pi x = \tan^{-1} (hE_n/2V)$. The resistance *R* must then be:

$$R = R_1 \frac{g}{h} \left[\frac{\frac{1}{4} - x^2}{x} \tan \pi x \right] \quad \dots (5)$$

If the factor between brackets is plotted as a function of *x*, a curve resembling a parabola with a flattened top is produced. A similar variation as a function of *x* is shown by the resistance between the output terminals of the circuit in fig. 9*b*. This contains a second potentiometer, *P*₂, whose wiper arm is coupled mechanically to that of *P*₁. The resistance of this circuit is:

$$R = d \frac{c + \frac{1}{4} - x^2}{c + d + \frac{1}{4} - x^2} R_2, \quad \dots (6)$$

where *c* and *d* are constants. This function does not exactly correspond to the function required, but the values of *R*₁, *R*₂, *c*, *d* and *h/g* may be chosen in such a way as to keep the relative error of the radius of curvature smaller than 3×10^{-5} over the whole range $|x| \leq \frac{1}{2}$ (this error is thus negligible compared with that caused by the normal non-linearity of the potentiometers *P*₁ and *P*₂). The distance *h* is fixed by the construction of the trolley, so that the value of the factor *g* can be calculated from *h/g*.

The servo amplifier

Since no current may flow in the circuit of the potentiometer tap, the voltage *V*_p is applied to an amplifier capable of delivering the power needed to energize the servo motor. The output voltage of this servo amplifier is an alternating voltage of 50 c/s, whose amplitude is proportional to that of *V*_p.

The waveform of *V*_p is shown in fig. 5. In the equilibrium position (the steering shaft is then correctly aligned) the middle of the plateau is at zero level (*b*, fig. 5). The servo amplifier is therefore required to amplify the voltage only during the mid-

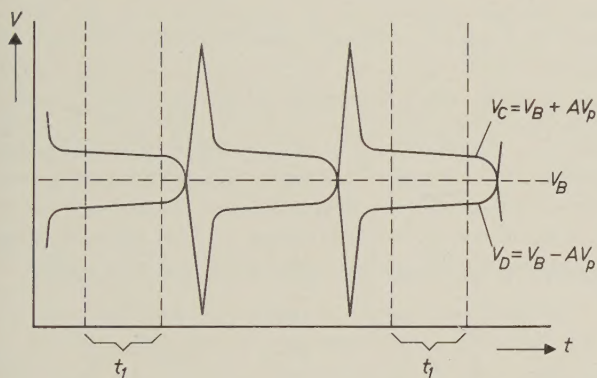
plateau part of each cycle (t_1 , see *fig. 10*). This is achieved in the following way.

The servo amplifier contains a stage which amplifies the voltage V_p to AV_p . This voltage is then added to a direct voltage V_B and also subtracted from it, thus producing two voltages

$$V_C = V_B + AV_p \quad \text{and}$$

$$V_D = V_B - AV_p$$

(*fig. 10*). V_C and V_D are fed to a gate circuit which, via the phase shifter FD (*fig. 3*), is controlled by the same alternating voltage that is applied, via the power amplifier, to the electrodes of the model. The phase difference between the two output voltages of FD is so adjusted that the voltages V_C and V_D are passed by the gate circuit to the next stage only during the intervals t_1 . This next stage is a modulator, which delivers an alternating voltage



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Fig. 10. Diagram of the voltages in the servo amplifier. The voltage V_p , after amplification, is added to V_B and also subtracted from it. During the time interval t_1 , these voltages V_C and V_D , respectively, are identical if the position of the steering shaft is correct.

of 50 c/s whose amplitude is proportional to the difference $V_C - V_D$, and thus proportional to the value of V_p during the interval t_1 . This also applies when V_p is negative, in which case the phase is shifted 180° .

The servo motor is a two-phase motor, one winding of which is fed via a transformer from the mains, the other being supplied with the output voltage from the servo amplifier. Depending on the phase of the latter voltage, the motor turns clockwise or anti-clockwise until V_p is zero.

It is now clear why the terminals of the power amplifier must not — as pointed out earlier — be interchanged. The phase of V_p would then be shifted 180° , and in the interval t_1 , therefore, V_p would have the opposite sign and the motor would rotate in the wrong direction.

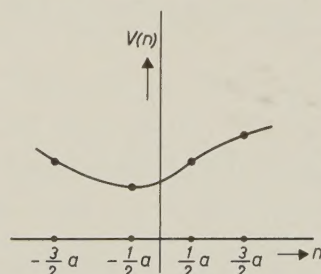
The entire servo system is required to respond

fast enough to enable the trolley to follow properly all the curvatures of the trajectory. This can be checked by, for example, letting the trajectory be plotted in both directions. Any discrepancy between the two trajectories indicates that the system has not responded rapidly enough, or that the trolley was moving too fast. The response time and the damping of the system depend on the moment of inertia of the rotating parts, on the friction, and on the ratio $M/\Delta a$, where M is the torque acting on the steering shaft as a result of a deviation Δa from its correct angular position. The response of the system is faster, and at the same time the damping smaller, the greater the value of $M/\Delta a$. The ratio $M/\Delta a$ is proportional to the gain of the servo amplifier and to the voltage V , and also depends to some extent on the radius of curvature. Since V is proportional to the electrode supply voltage, $M/\Delta a$ can be adjusted either with the servo amplifier or with the power amplifier.

The system works best, that is to say the trolley speed can be greatest, when there is hardly any damping and the amplitude of the trace remains small. The trolley speed can be about 2 cm/sec if the trajectory curvature radius ϱ varies only slightly, but it must be substantially less if the variations of ϱ are considerable. For this purpose the supply voltage of the drive motor can be varied with a potentiometer whilst the trajectory is being plotted.

Derivation of the formulae for E_n and V

The field strength E_n and the potential V must be derived by the computing circuits from the voltages of the four probes. We call the line through the probes, which is perpendicular to the direction of the trajectory, the n axis (*fig. 11*). The n -coordinates of the probes $S_1 \dots S_4$ are: $-\frac{3}{2}a$, $-\frac{1}{2}a$, $\frac{1}{2}a$ and $\frac{3}{2}a$; the distance between the probes is a . Four values are known of the function $V(n)$ that gives the potential variation along the n axis. We assume that $V(n)$



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Fig. 11. Potential distribution along the n axis. The probes are situated at points $n = -\frac{3}{2}a$, $n = -\frac{1}{2}a$, etc. The function $V(n)$, which describes the potential variation, is approximated by a third-degree curve through the points representing the potentials of the four probes.

is a third-degree polynomial in n , for this is the most general polynomial determined by four values. We can now calculate the function $V(n)$, from which $V = V(0)$ and $E_n = -V'(0)$ can be evaluated⁵⁾. We find:

$$E_n = \frac{1}{24a} [27(V_2 - V_3) + (V_4 - V_1)] \quad (7)$$

$$\text{and } V = \frac{1}{16} [9(V_2 + V_3) - (V_1 + V_4)] \quad (8)$$

The effect of induced dipoles

A complication arises in that the probes disturb the potential field in the electrolyte. In each probe a dipole is induced which affects the field near the other probes. The voltages $V_1 \dots V_4$ measured on the probes are consequently not the same as the voltages $V_{01} \dots V_{04}$ prevailing in the electrolyte when the probes have not yet been immersed. The latter voltages must be used instead of $V_1 \dots V_4$ in formulae (7) and (8), and therefore we must ascertain how these voltages are derived from the measured $V_1 \dots V_4$.

The dipole moments of the four probes are proportional to the local field strengths and have the same direction. Since the components perpendicular to the n direction have no influence on the field near the other probes, we shall consider only the components in the n direction, $M_1 \dots M_4$. These are proportional to the n components of the local field strengths, $E_1 \dots E_4$:

$$M_i = \varepsilon k E_i \quad (i = 1 \dots 4), \quad (9)$$

where ε is the dielectric constant of the electrolyte, and the factor k has the dimension (length)³.

Between the voltages V_{0i} and V_i the following relations exist:

$$\left. \begin{aligned} V_{01} &= V_1 + \frac{k}{(6a)^3} (71V_1 + 126V_2 - 171V_3 - 26V_4), \\ V_{02} &= V_2 + \frac{k}{(6a)^3} (-414V_1 + 783V_2 - 270V_3 - 99V_4), \\ V_{03} &= V_3 + \frac{k}{(6a)^3} (-99V_1 + 270V_2 - 783V_3 - 414V_4), \\ V_{04} &= V_4 + \frac{k}{(6a)^3} (-26V_1 + 171V_2 + 126V_3 - 71V_4). \end{aligned} \right\} \quad (10)$$

In formula (7) for E_n we must replace $V_1 \dots V_4$ by $V_{01} \dots V_{04}$. Substituting the expressions (10) for $V_{01} \dots V_{04}$ we obtain:

$$E_n = \frac{1}{24a} \left[27(V_2 - V_3) + (V_4 - V_1) + \frac{k}{a^3} \{ 130(V_2 - V_3) + 40(V_4 - V_1) \} \right] \quad (11)$$

The same manipulations in respect of formula (8) for V results in:

$$V = \frac{1}{2}(V_2 + V_3) + \frac{1}{16}(V_2 + V_3 - V_1 - V_4)(1 + 21.6 \frac{k}{a^3}) \quad (12)$$

The second term is already small in relation to the first, so there is no point in applying the correction factor to this term. As regards the voltage V , therefore, the correction for the dipole moments of the probes need not be applied and (12) reduces to equation (8).

The equations in (10) are derived from various formulae of electrostatics, confined in the present case to the above-mentioned components in the n direction and to points on the n axis.

If a dipole, the n component of whose moment is M , is situated at the origin of the n axis, the field strength at the point $n = a$ is given by $E = 2M/|a|^3$ and the potential at that point by $V = Ma/|a|^3$. Bearing in mind that the field near a probe is built up from the original field and the contribution from the dipoles of the other probes, we find, where E_{0i} is the n component of the field strength for non-immersed probes:

$$\left. \begin{aligned} E_1 &= E_{01} + \frac{2k}{a^3} E_2 + \frac{2k}{(2a)^3} E_3 + \frac{2k}{(3a)^3} E_4, \\ E_2 &= E_{02} + \frac{2k}{a^3} E_1 + \frac{2k}{a^3} E_3 + \frac{2k}{(2a)^3} E_4, \end{aligned} \right\} \quad (13)$$

$$\left. \begin{aligned} V_1 &= V_{01} - \frac{k}{a^2} E_2 - \frac{k}{(2a)^2} E_3 - \frac{k}{(3a)^2} E_4, \\ V_2 &= V_{02} + \frac{k}{a^2} E_1 - \frac{k}{a^2} E_3 - \frac{k}{(2a)^2} E_4. \end{aligned} \right\} \quad (14)$$

Similar relations hold for E_3, E_4, V_3 and V_4 .

We further assume, for the reason already mentioned, that the function giving the potential variation $V_0(n)$ along the n axis when the probes are not immersed in the water, is a third-degree polynomial in n . The first derivatives at points $-\frac{3}{2}a, -\frac{1}{2}a, \frac{1}{2}a$ and $\frac{3}{2}a$ are the field strengths $E_{01} \dots E_{04}$. This gives another four equations between the quantities. We now have 12 equations available, containing 16 variables. After elimination (matrix algebra is the simplest way of doing this) we can express $V_{01} \dots V_{04}$ in terms of $V_1 \dots V_4$, which yields the above result.

The value of k/a^3 can be calculated from the formula for the dipole moment M induced in a cylinder of radius R and height h , where the field strength E is normal to the axis: $M = \frac{1}{3}\varepsilon R^2 h E$. Comparison with (9) yields:

$$k = \frac{1}{3} R^2 h.$$

The diameter of the probes is 0.5 mm and they project about 1 mm into the electrolyte. The distance a is 4 mm, and hence $k/a^3 \approx 0.0005$.

⁵⁾ For a more detailed treatment see: J. L. Verster, On the use of gauzes in electron optics, dissertation Delft, to be published shortly by Centrex Publishing Co., Eindhoven.

The computing circuit for E_n

It follows from (11) that E_n is given by:

$$E_n = \frac{1}{24a} \left[(27 + 130 \frac{k}{a^3})(V_2 - V_3) + (1 + 40 \frac{k}{a^3})(V_4 - V_1) \right]. \quad (15)$$

The circuit required to deliver a voltage proportional to E_n must be a kind of difference amplifier. The output voltage V_o of an ideal difference amplifier is proportional to the difference of two input voltages V_{i1} and V_{i2} . It is difficult, however, to make a difference amplifier whose output voltage is not at the same time to some extent dependent on $V_{i1} + V_{i2}$. In that case:

$$V_o = A \left\{ (V_{i1} - V_{i2}) + \frac{1}{H} (V_{i1} + V_{i2}) \right\}, \quad (16)$$

where H is the so-called rejection factor⁶⁾. For an ideal difference amplifier, $H = \infty$. The second term in eq. (16) can still be minimized, however, by ensuring that $V_{i1} + V_{i2}$ is small. To this end, a voltage is deducted from both input voltages which has roughly the mean value $\frac{1}{2}(V_{i1} + V_{i2})$.

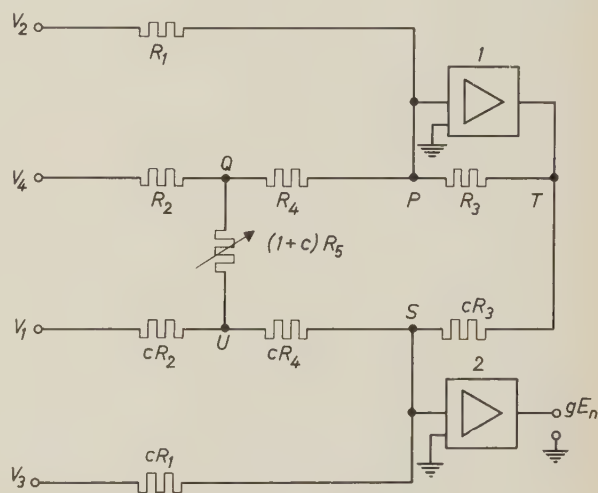
This operation is done on the input voltage to the difference amplifier for computing E_n . It is for this reason, as mentioned on page 250, that the voltage V is subtracted from the voltages $V_1 \dots V_4$, the value of V being close to the average voltages for both subtractions in eq. (15). A further incidental advantage of this method is that it considerably reduces the influence of differences between the cathode followers, which might give rise to the same error as a finite rejection factor.

The principle of the circuit for computing E_n is illustrated in fig. 12. Amplifier 1 has a high gain, so that the potential at point P is zero. The resistances of the network are chosen such that V_S at point S is proportional to E_n , and only R_5 is dependent on the constants k and a of the apparatus. No current may be drawn from point S , and for this reason amplifier 2 is of a type that has a high input impedance and whose output may therefore permissibly be loaded. The gain is adjusted to give the output voltage gE_n .

High demands are made on the accuracy of the computing circuit, which means that strong negative feedback is required from amplifier 1. In view

of the high gain which the latter must have, this makes it difficult to achieve a stable circuit. A solution was found by utilizing a new principle termed "addition of the complement"⁷⁾. A main amplifier raises the input signal to roughly the required output signal. A combination of input and output signal is amplified by a correction amplifier, and this combination vanishes when the output signal has the correct value. The output signal from the correction amplifier is added to that of the main amplifier. With this arrangement an accurately defined gain is obtained, without the gain of the individual amplifiers having to be particularly high. The negative feedback in these amplifiers is relatively small, so that no complicated feedback network is needed to make the circuit stable.

Since the gain of amplifier 2 need not be high (about 60), it is sufficient to use a conventional amplifier consisting of two pentodes with negative current feedback to the cathode of the first tube.



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Fig. 12. Block diagram of the computing circuit for E_n . The probe potentials are applied to the terminals on the left. Amplifier 1 has a high gain, so that the potential of point P is effectively zero. The resistance values are such as to make V_S proportional to E_n . Since the network must not carry current, amplifier 2 is given a high input impedance. Only R_5 is dependent on the dimensions of the probes.

The computing circuit for V

The computing circuit for V is required to supply the voltages V and $-V$ to the trolley. At the same time the voltage $-V$ must be applied to the "cathode" output of the power amplifier in order to make the potential at the point midway between the probes equal to zero. We shall explain how the latter is achieved in this way by considering a

⁶⁾ See G. Klein, Rejection factor of difference amplifiers, Philips Res. Repts. 10, 241-259, 1955; see also Philips tech. Rev. 21, 32-33, 1959/60.

⁷⁾ J. J. Zaalberg van Zelst, Stabilised amplifiers, Philips tech. Rev. 9, 25-32, 1947/48.

simple case as represented in *fig. 13*. Here there is only one probe, which has a potential V relative to the cathode. If V_k is the potential of the cathode

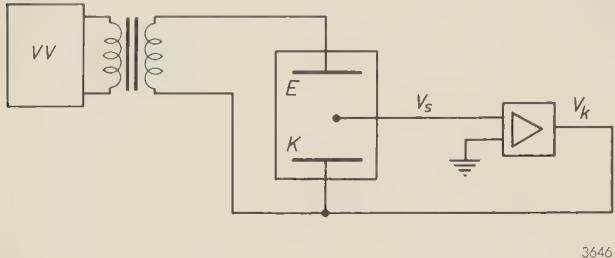


Fig. 13. Circuit for keeping the probe at earth potential, for the case of only one probe. VV power amplifier for supplying the electrodes E and K (K is the cathode of the system). The voltage between the probe and the cathode is V . If the gain A is very much greater than unity, $V_s = 0$ and $V_k = -V$.

relative to earth, the probe voltage with respect to earth is:

$$V_s = V_k + V. \quad (17)$$

The figure shows further that:

$$V_k = -AV_s. \quad (18)$$

It follows from eq. (17) and (18) that:

$$V_k = -\frac{V}{1 + 1/A} \quad \text{and} \quad V_s = \frac{V}{A + 1}.$$

If the gain A is sufficiently high, we can thus put $V_s = 0$ and $V_k = -V$. The voltage V can be obtained by using an inverting stage.

When four probes are used, the output voltage must satisfy eq. (8). The principle of the circuit used in this case is illustrated in *fig. 14*. For the input voltage V_{p1} a voltage divider of high resistance is used to obtain the mean of V_2 and V_3 , so that V_{p1} with respect to earth is:

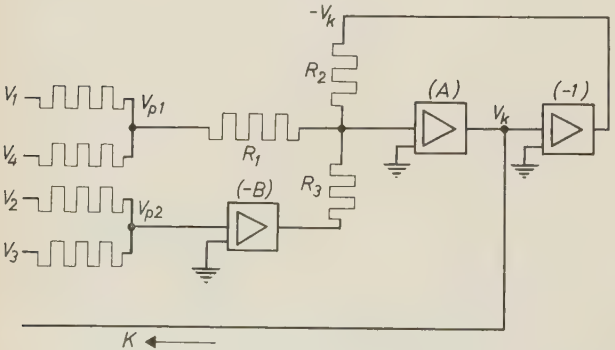


Fig. 14. Principle of the computing circuit for V . This circuit ensures that the potential at the point midway between the probes always remains zero. The resistances and gain figures A and B are chosen such that $V_k = -V$; the inverting amplifier delivers the voltage V .

$$V_{p1} = \frac{1}{2}(V_1 + V_4) + V_k. \quad (19)$$

Likewise:

$$V_{p2} = \frac{1}{2}(V_2 + V_3) + V_k. \quad (20)$$

By a suitable choice of the resistances R_1 , R_2 and R_3 and the gains A and B , we can obtain (see eq. (8)):

$$V_k = -\frac{9}{16}(V_2 + V_3) + \frac{1}{16}(V_1 + V_4) = -V. \quad (21)$$

As the cathode now has a potential $-V$, the potential midway between the probes is zero. The circuit produces both V and $-V$. The gains A and B need not be high, so that simple amplifiers can be used. The inverting amplifier is again designed on the principle of "addition of the complement".

The cathode followers

The probes, regarded as voltage sources, have a high internal resistance owing to the fact that the part projecting into the electrolyte is small and the conductivity of the electrolyte is low. To prevent the probes drawing current from the electrolyte, they are connected to cathode followers which pass the voltages on to the computing circuits.

These cathode followers must meet three requirements:

- a) The input impedance must be high compared with the probe impedance.
- b) The ratio of the output voltage V_o to the input voltage V_i must be highly constant and identical for all cathode followers.
- c) The output impedance must be small compared with the input impedance of the two computing circuits in parallel.

The latter requirement is necessary because the cathode followers are not equally loaded by the computing circuits. A cathode follower consisting of one valve does not simultaneously fulfil the second and third requirements. If the output impedance is made small, the output voltage is then too dependent on the valve characteristics and therefore can no longer be identical for all four. A cascade arrangement of two valves (*fig. 15*) satisfies the above conditions (see reference ⁵) for the calculation), provided the following measures are taken:

- a) To achieve a high input impedance, the grid of valve 2 is directly coupled with the probe, thus dispensing with the need for a grid leak. This grid now receives, in addition to the alternating voltage, the direct voltage from the probe, which is approximately zero.
- b) To minimize the dependence of V_o/V_i on the valve characteristics, the amplification factor of valve 2 must be high. To minimize the load, the inter-

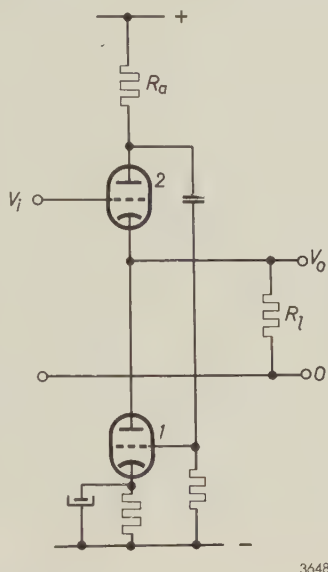
nal resistance of valve 1 must be high. Both valves used are therefore pentodes.

- c) A low output impedance is achieved by using valves having a fairly steep slope and large R_a . Measurements yielded the following values for all cathode followers:

$$V_o/V_i = 0.9998 \pm 0.0001 \text{ where } R_l = \infty,$$

$$V_o/V_i = 0.9996 \pm 0.0001 \text{ where } R_l = 4.7 \text{ k}\Omega.$$

Fig. 15. Basic diagram of a cathode follower consisting of two valves in cascade. If the two valves are pentodes, the gain differs very little from unity and is largely independent of the valve characteristics. Moreover, the input impedance is very high since no grid leak is needed, the cathode follower being directly coupled to its probe and thus biased to the potential of the probe, which differs little from zero.



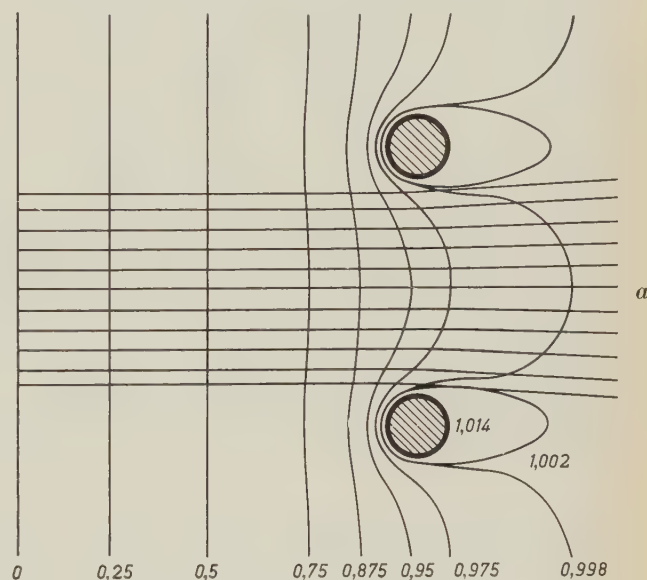
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Applications and performance

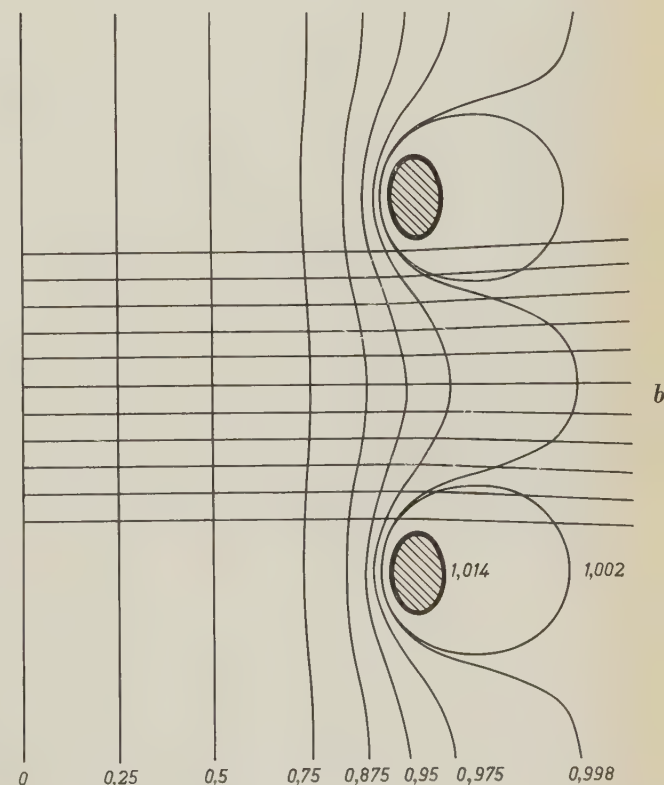
In electrostatic fields with no space charge the apparatus described is capable of plotting electron trajectories lying in planes of symmetry. For electrostatic lenses having rotational symmetry, these are the planes through the axis. By plotting a series of trajectories from different starting points and with different initial directions, it is possible to determine the focal planes, principal planes, spherical aberration, distortion and field curvature of such lenses. Certain forms of misalignment and axial astigmatism (lens errors caused by the electrodes not being centered on the axis or being non-circular) can also be investigated.

As our first example we shall consider the trajectories in a planar triode having an intersecting-“wire” gauze grid, where the grid is at a positive

potential, and the average field strength between grid and anode is zero. Fig. 16a shows the trajectories in a perpendicular section which cuts the gauze parallel and perpendicular to the directions of the “wires”, and fig. 16b in a section that cuts the gauze diagonally. We see that in the first case the trajectories become more divergent the closer they pass by the grid “wires”. In the second case the trajectories are again divergent, but their divergence shows a maximum, that is to say the trajectories passing



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Fig. 16. The electron trajectories and equipotential lines in the field of a planar triode with an intersecting-“wire” gauze grid. (These are drawings copied from the plotted trajectories.) a) The plane in which the trajectories are drawn cuts the gauze parallel and perpendicular to the directions of the “wires”. b) The plane cuts the gauze diagonally.

In both drawings the cathode, which has zero potential, is on the left, and the grid, which has a positive potential, is on the right. The anode is not shown. Between grid and anode there exists an equipotential space. The potential of this space is put equal to 1, and the relative potential with respect to this value is marked beside the equipotential lines.

close by the points of intersection of the grid wires are deflected less than those that pass somewhat farther away. Also drawn in fig. 16a and b are the equipotential lines.

The second example (fig. 17a) concerns the

electron trajectories in an accelerating lens consisting of two adjacent coaxial cylinders of identical diameter. Both cylinders have a positive potential with respect to the cathode, the right one being 20 times as high as the left one. The trajectories drawn

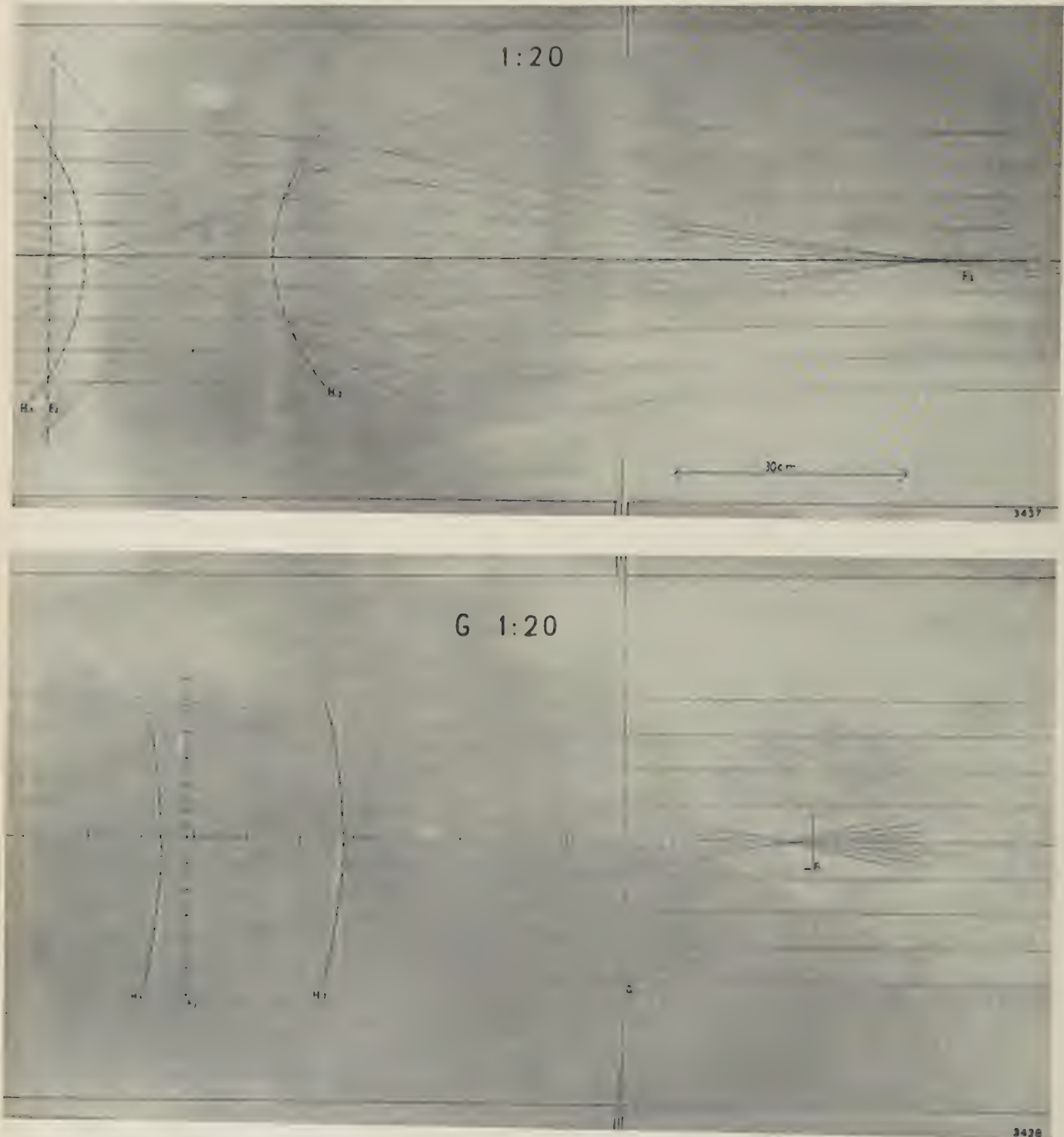


Fig. 17. a) Electron trajectories as traced out by the automatic plotter for the model of an accelerating lens. (The electrodes of this model are shown in fig. 1.) The lens consists of two adjacent coaxial cylinders of the same diameter; the potential with respect to earth of the right cylinder is 20 times that of the left one. The trajectories are drawn in a plane through the axis of the lens. The electrons enter parallel to the axis, from the left and also from the right. The principal planes and focal planes are indicated, together with the situation of the electrodes.
b) The trajectories in the same accelerating lens, but now with a plane gauze G at the inner end of the right cylinder, having the same potential as the cylinder. It can be seen that this makes the lens more powerful and results in a considerably smaller curvature of the focal planes.

are those of electrons entering parallel to the axis, both from the left and from the right. From these the focal planes and principal planes can be determined. Fig. 17b shows the trajectories in the same lens when a flat gauze is fixed on the inner end of the right cylinder, the gauze having the same potential as the cylinder. It can be seen that the power of the lens is now considerably greater. (For a detailed treatment, see reference ⁵.)

Potential distributions can also be measured with this instrument. The determination of a potential at a particular point in a field is greatly simplified by making use of various components that are also used for plotting electron trajectories. The ends of the decade potentiometer P_{dec} are then connected to the electrodes which are respectively at the lowest and highest potential. The voltage on the wiper of P_{dec} is applied to an oscilloscope. The stylus on the trolley is placed over the measuring point, and P_{dec} is adjusted until the voltage on its wiper is zero. Since the computing circuit for V keeps the potential at the point midway between the probes equal to zero, the wiper of P_{dec} and the measuring point now have the same potential with respect to the electrodes. This potential can be derived from the position of P_{dec} .

In order to plot a complete equipotential line, P_{dec} is again connected up in the same way and its wiper is adjusted to the potential of the equipotential line in question. When the stylus is above a point that possesses this potential with respect to the electrodes, the voltage of the wiper with respect to earth will then, for the same reason as above, be zero. The circuit is now modified as follows. The steering circuit and the servo motor are disconnected, and the wiper of P_{dec} is connected to the input of the servo amplifier. The output of this amplifier energizes a relay. A make contact of this relay is connected in parallel with the stylus relay, so that the latter is energized only when the voltage from the servo

amplifier, and thus the potential of the wiper, drops to zero. When the trolley is now moved to and fro by hand over the place where the equipotential line is presumed to be, successive dashes are recorded. These dashes are later joined up to make a continuous line.

The automatic plotter has an accuracy of 0.2%, which means that the relative error in the radius of curvature of the trajectory is smaller at every point than 0.2%. All circuits and components of the plotter must be optimally adjusted, and the speed of the trolley must be properly adapted to the curvature of the path described. The average time taken to plot a trajectory is about 2 to 3 minutes per meter.

The accuracy of the system should be checked now and then by plotting a number of paths in the field between two parallel plates (the paths should be parabola) and in the field of a spherical capacitor. In both these cases the form of the paths can be derived exactly. Any components or circuits requiring re-adjustment can be identified from the deviations shown by the paths thus described.

Summary. Electron trajectories in an electrostatic field can be automatically plotted by an apparatus designed round an electrolytic tank. The tank contains a model of the electrode system in which the trajectories are to be determined. On a board above the tank rides a three-wheel trolley which is mechanically coupled to four closely-spaced probes, mounted in line and dipping in the electrolyte. The radius of curvature of the trajectory at the point midway between the probes is derived from the voltages of the four probes. The apparatus does this by means of two computing circuits. A servo system ensures that the trolley — propelled at a suitable speed of, say, half a meter per minute — describes a path which everywhere has the correct radius of curvature. The trajectory is traced by a stylus fixed underneath the trolley.

The electrodes of the model are fed with a square-wave alternating voltage of 500 c/s with the object of minimizing polarization. For the same purpose the electrodes are silver-plated. The accuracy achieved is 0.2%. Practical examples mentioned are the plotting of electron trajectories in an accelerating lens, with and without a gauze, and the trajectories in a planar triode. The instrument can also be used as a particularly simple means of determining equipotential lines.

A SMALL GETTER ION-PUMP

by A. KLOPFER *) and W. ERMERICH *).

621.528.5/.6

The advances made in vacuum technique in the last ten years have given a new lease of life to the apparently outmoded idea of continuously pumping a vacuum tube during operation.

The getter ion-pump forms a compact, convenient vacuum pump, immediately set in operation and capable of pumping small quantities of gas to maintain a low pressure for a long period of time.

The principle of this pump has been known for some years ¹⁾: a getter material, such as zirconium or titanium, is continuously or intermittently evaporated, and the gas molecules are ionized in the same way as in a hot-cathode ionization gauge ²⁾ or a Penning vacuum gauge ³⁾. The ions formed are then trapped in the getter film deposited on the inside of the envelope.

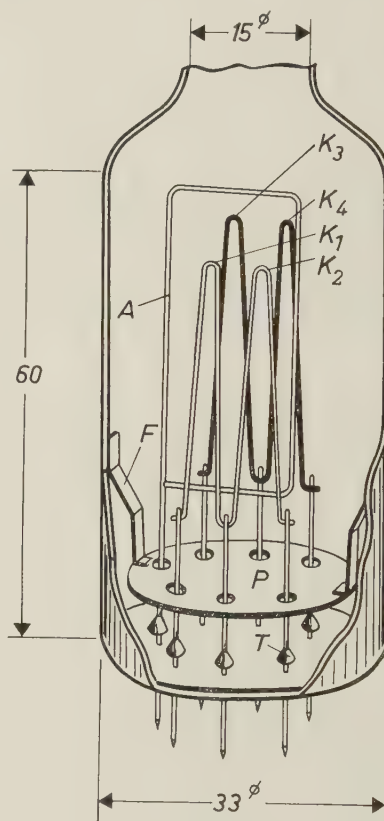
In this article a getter ion-pump will be described in which a certain quantity of titanium is evaporated, and where the titanium vapour itself simultaneously sustains a discharge of the type occurring in a Penning vacuum gauge. In conclusion, various examples of the pump's application will be discussed.

Construction and operation of the getter ion-pump

The pump consists of a glass envelope containing an electrode system. The electrode assembly (fig. 1) is similar to that in a Penning gauge. The Penning system is formed by the loop-shaped anode *A*, made of molybdenum, the cathodes *K*₁-*K*₄, and a magnetic field. The cathodes carry a supply of getter material. The direction of the magnetic field, with an induction of 0.04 Wb/m², is perpendicular to the plane of the anode. The electrodes are mounted on a glass base. The cylindrical glass bulb measures 33 mm in diameter and 60 mm in length. The weight of the pump (40 g) together with the permanent magnet is 450 g. The complete assembly is therefore conveniently light and easy to connect up with the object to be evacuated.

The pump is set in operation by applying a DC potential of 2 kV between anode and cathodes, the latter being heated by an alternating current. It is

also possible to heat only one cathode. The cathodes consist of stranded tungsten and titanium wires, and are heated to a temperature at which the titanium evaporates. The rate of evaporation is governed by the temperature and the number of cathodes



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Fig. 1. Getter ion-pump in glass envelope. *K*₁-*K*₄ cathodes. *A* anode. *P* shield. *T* protective caps. *F* contact spring.

heated. Part of the evaporated getter material settles directly on the glass walls of the pump; another part contributes to the Penning discharge, in that, during evaporation, titanium atoms are ionized and electrons are released. If the total pressure of the gases present is less than 10⁻³ torr (10⁻³ mm Hg), the discharge is sustained not by these gases but almost entirely by the titanium vapour. The molybdenum shield *P*, together with the protective caps *T*, serves to prevent short-circuiting between anode and cathode if the titanium were to

*) Philips Zentrallaboratorium GmbH, Aachen Laboratory.

¹⁾ R. G. Herb, R. H. Davis, A. S. Divatia and D. Saxon, Phys. Rev. **89**, 897, 1953.

²⁾ A. Venema and M. Bandringa, The production and measurement of ultra-high vacua, Philips tech. Rev. **20**, 145-157, 1958/59.

³⁾ F. M. Penning, Physica **4**, 71, 1937; see also Philips tech. Rev. **11**, 116, 1949/50.

deposit on the glass base between the lead-ins. That can only happen, however, if the titanium is evaporated at excessive pressures, causing the mean free path of the titanium atoms to become shorter than the distance to the glass wall. In that case, as a result of collisions in the gas, titanium atoms may arrive on the base. If the pump is to be used for producing very high vacua, so that the titanium is only evaporated at low pressures, the shield *P* can better be removed. The reason for this is that, for such low pressures, all metal parts must be degassed by heating; this can be done very simply for the electrodes by passing a current through them, but this is not practicable in the case of the shield. (The anode loop would be led out through the base via two pins to enable current to be passed through it.)

The power supply for the pump consists merely of a DC voltage source of 2 kV, 2 mA, and an AC voltage source of 10 V, 10 A.

The pump operates as follows.

- 1) The getter material deposited on the glass wall adsorbs the molecules of the chemically active gases impinging on the surface, such as oxygen, hydrogen, nitrogen, etc.
- 2) The Penning discharge contains a vast number of ions and electrons. The ions, but especially the electrons, which can describe a long path owing to the presence of the magnetic field, have a considerable chance of colliding with gas molecules entering the discharge from the space to be evacuated. The chemically active as well as the inert gas molecules are thereby ionized. The gas ions formed are then, under the action of the electric field, shot into the getter film — which is kept at cathode potential by the contact spring *F* — and there they are trapped.

As a result of continuously evaporating the titanium, the getter ion-pump, as its name implies, has a double action in that a fresh surface is continuously created for trapping ions, and a fresh chemically active surface for gettering.

Properties of the pump

The characteristic properties determining the performance of a getter ion-pump are:

- a) the maximum permissible initial pressure,
- b) the pumping capacity,
- c) the pumping speed, and
- d) the lowest attainable pressure (ultimate pressure).

The maximum permissible initial pressure, which is determined by the method of evaporation chosen, amounts in the pump described here to a few tenths of a torr.

The pumping capacity for chemically active

gases, such as CO and H₂, i.e. the total quantity that can be extracted, is given by the getter supply present in the cathode. For chemically active gases the pumping capacity is equal to the gettering capacity. This differs according to the gas, the maximum value being achieved when each getter atom traps as many gas molecules as corresponds to the chemical reaction equation. In practice this situation seldom obtains. For each cathode we generally use 12.5 mg of titanium in the form of three wires, each of 100 microns diameter, stranded with the tungsten heater wire. The four cathodes together thus contain 50 mg of titanium. The quantity of gas that can be removed under favourable conditions with this quantity of getter material is about 2.5 torr.litre in the case of CO, calculated for a maximum specific gettering capacity of 0.05 torr.litre/mg. Since a vacuum system may contain a mixture of the most diverse chemically active gases, one can generally reckon only with an average specific gettering capacity of 0.01 torr.litre/mg.

For inert gases, such as He and CH₄, the pumping capacity is considerably less. Measurements have shown, however, that in the case of He it is certainly more than 0.05 torr.litre.

In principle the cathodes could accommodate a larger supply of getter material.

The quantity of gas removed per second in the steady state is given for a chemically active gas by the expression:

$$\frac{dQ}{dt} = C \frac{dM}{dt} = Sp. \quad (1)$$

Here dM/dt represents the rate of evaporation of the getter material, *C* the specific gettering capacity, *S* the pumping speed (governed by the speed with which the getter takes up gas and the geometry of the glass envelope of the pump) and *p* the resultant pressure at the pump mouth, i.e. the place where the getter ion-pump is connected to the vessel to be evacuated. Fig. 2 shows the measured pumping speeds *S* in l/sec as a function of the rate of evaporation dM/dt in mg/sec, for various quantities of CO supplied per unit time in torr.litre/sec. The maximum value for *S*, in this case 62 l/sec, is determined by the orifice of the pump mouth. For other chemically active gases the pumping speed *S* is of the same order of magnitude. The curves in fig. 2 were obtained irrespective of whether the anode was under high tension or not. This means that, for CO, the contribution of ion trapping to the total pumping speed in the measuring range investigated is small compared with the contribution of the gettering action alone.

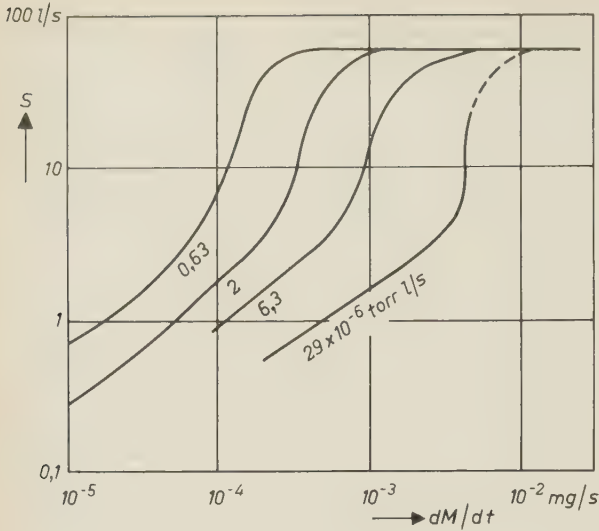


Fig. 2. Pumping speed S in l/sec as a function of evaporation rate dM/dt in mg/sec, for various influx rates of CO in torr.litre/sec.

In order to use equation (1) it is desirable to determine the relation between the specific gettering capacity C and the pumping speed S . From the curves in fig. 2 we can calculate for any value of S the corresponding values of C for the various quantities of CO supplied. The surprising result is that the same value of C is always found for a given S . A plot of S versus C is shown in fig. 3. Along the abscissa at the top the specific gettering capacity is set forth in torr.litre/mg, and at the bottom the quantity C as the ratio between the number of CO molecules trapped

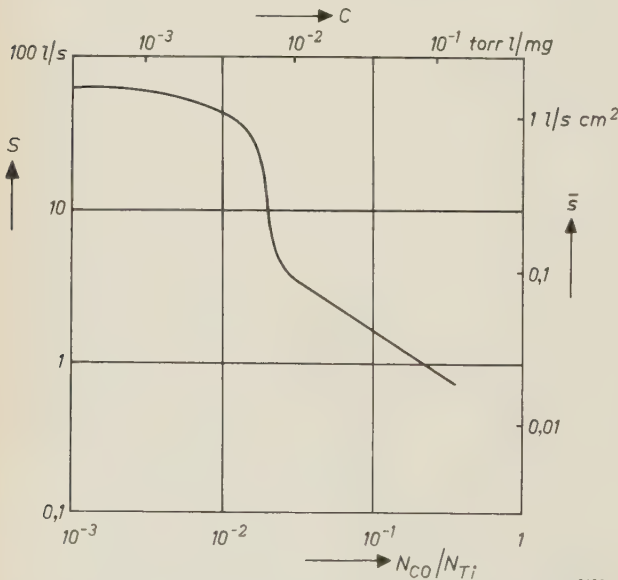


Fig. 3. Pumping speed S in l/sec as a function of specific gettering capacity C in torr.litre/mg, calculated from the data in fig. 2; or, which is equivalent, the ratio N_{CO}/N_{Ti} between the number of gettered CO molecules and the number of titanium atoms incident on the getter surface, as a function of $\bar{S}$, the mean pumping speed (in l/sec) of the getter film per cm².

and the number of titanium atoms incident on the getter surface. Only at large values of S is the relation between S and C no longer certain, for it is here, as we have seen, that the size of the pump orifice imposes an upper limit on the pumping speed. Apart from this, the pumping speed is established for any given value of C . We may therefore deduce that a constant S , independent of the influx rate of gas, can only be obtained if the rate of evaporation varies in proportion to the pressure in the pump, since it follows from (1) that

$$S = C \left(\frac{dM}{dt} p \right) \dots \dots (2)$$

A fairly good approximation to the rate of evaporation of the hot cathode is arrived at by deducting from the discharge current I in the pump the discharge current when the cathode is cold. The relation between the rate of evaporation and the discharge current is represented by the curve in fig. 4 (corrected for residual gas).

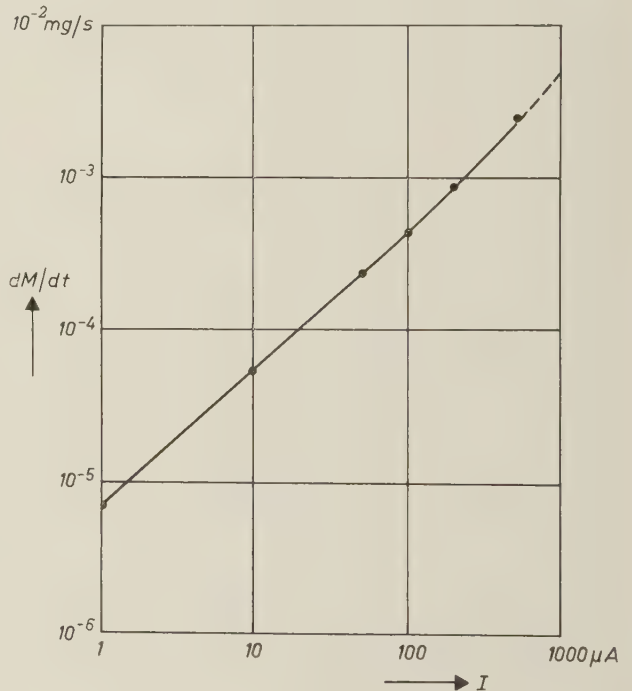


Fig. 4. Evaporation rate dM/dt in mg/sec of one cathode in the getter ion-pump, as a function of the discharge current I in μA .

The above considerations make it evident that the discharge current when the cathode is hot is no measure of the gas pressure. It is, however, when the cathode is cold, and — which is sometimes convenient — the pump may then be used for pressure measurements in a certain range of pressures, as shown by the curves in fig. 5. These curves relate to both N_2 and CO at a magnetic induction of 0.04 Wb/m² and an anode voltage of 1 and 2 kV, respectively, with an anode resistor of 1 M Ω .

The pumping speeds found for chemically inactive gases, such as the rare gases and methane, are much

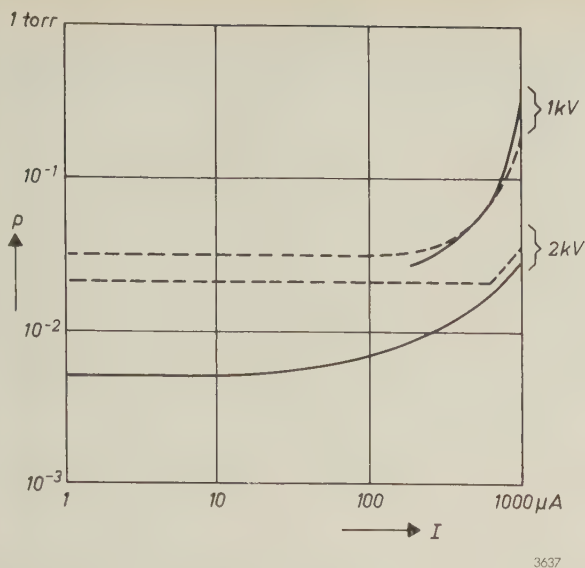


Fig. 5. The getter ion-pump as a pressure gauge: the pressure p in torr as a function of the discharge current I in μA , with cold cathode, for two anode voltages. Full curves: with getter film; dashed curves: without getter film.

lower (see Table I). It can be seen from the table that the pumping speed varies only slightly with the discharge current. The rare gases are not chemically bound by the layer of evaporated titanium, and in this case the pumping action relies entirely on the ionization in the gas. The ions formed are shot into the titanium layer and trapped there. How long they remain trapped will depend entirely on the rate of diffusion in the titanium layer. As the rate of diffusion is temperature-dependent, it is advisable to cool the pump wall with a fan. Where methane is concerned, thermal decomposition at the hot cathode also makes an essential contribution to the pumping speed.

The lowest pressure obtainable with this getter ion-pump depends very much on the way in which the pump itself and the vessel are degassed. Degassing is necessary for various reasons, mainly to reduce the hydrogen content of the getter reserve and to prevent the unwanted formation of non-active gases such as CH_4 as a result of reactions between getter and gas ⁴⁾. The formation of chemically inactive gases is undesirable because of the low pumping speed for such gases.

On a small vacuum system having a volume of 0.5 l, which consisted of an omegatron, an ion gauge of the Bayard-Alpert type and a getter ion-pump of the type here described, we measured with the omegatron the partial pressures of the various gas components during the process of evacuation. Four such systems, in various degassed states, were investigated.

⁴⁾ A. Klopfer and W. Ermrich, *Vakuum-Technik* **8**, 162, 1959.

A system which had not been degassed at all was found to contain, after evaporation of the getter, mainly water vapour, carbon dioxide and hydrocarbons. The pressure of H_2O and CO_2 is governed by the release of gas from the glass surfaces and by the effective pumping speed at the position of the omegatron. The presence of hydrocarbons is attributable to two reactions: in the first place, during the evaporation, hydrogen and carbon present as impurities in the getter may react with one another and form hydrocarbons with up to five carbon atoms. In the second place, hydrocarbons may be formed from water and carbon at temperatures as low as room temperature, as we were able to demonstrate. As a result of the above-mentioned gas desorption and gas reactions, the lowest pressure in a system that has not been degassed lies between 10^{-7} and 10^{-8} torr ⁵⁾.

Table I. Pumping speed S for various non-active gases.

Gas	Discharge current I μA	Pumping speed S 10^{-3} l/sec	Pressure p 10^{-3} torr
He	100-1000	2	0.01-0.03
Ne	500	5.5	~ 1
Ar	100	3.5	~ 1
	500	7.5	~ 1
CH_4	100	74	~ 1
	500	210	~ 1

Even slight preliminary degassing causes a marked drop in the release of H_2O and CO_2 . The pumping time and the final pressure attainable are then entirely governed by the formation of hydrocarbons from H_2 and C during the evaporation of the getter, and by the pumping speed of the ion pump in respect of hydrocarbons. The pumping times are shorter the more thoroughly the vacuum system and the getter wires have been degassed. Pressures of about 1×10^{-9} torr can be achieved after relatively little preliminary degassing. The residual gas is principally composed of CH_4 , CO and H_2 . Thorough degassing, as described by Alpert ⁶⁾, makes it possible to reach extremely low pressures ($< 10^{-10}$ torr). The final pressure is then determined by the equilibrium pressure of the hydrogen dissolved in the titanium, and by the diffusion of atmospheric helium through the glass wall ⁴⁾⁷⁾. Heating of the getter film by the hot cathode promotes the formation of CH_4 and raises the equilibrium pressure of the hydrogen.

⁵⁾ G. Reich and H. G. Nöller, *Vacuum Tech. Trans.* **4**, 97, 1959.

⁶⁾ D. Alpert and R. S. Buritz, *J. appl. Phys.* **25**, 202, 1954.

⁷⁾ F. Norton, *J. appl. Phys.* **28**, 34, 1957.

For this reason too it is advisable to cool the pump wall with a fan.

Some applications

In order to produce vacuum required in electron tubes, the tubes are subjected during manufacture to two different evacuation operations. First of all, the electron tube is degassed and evacuated at suitable temperatures for a sufficiently long period with the aid of a good high-vacuum pump, usually a diffusion pump. Because of the bulk of the pumping equipment, the tube during this period is virtually immovable. At the end of this major evacuation process, the system consisting of electron tube and getter is sealed off. Further degassing can be effected by letting the tube operate normally or by even overloading it, whereby the evaporated getter material together with the ionizing electron current in the tube is responsible for reducing the residual gas pressure. The loading of the tube must not be increased so fast as to cause the pressure to assume values at which components, e.g. a photo-cathode, might be damaged.

The use of the small getter ion-pump described has considerable advantages for the evacuation of special electron tubes, the degassing of which must often be very protracted. For example, the outgassing times during evacuation by the diffusion pump can be shortened, and the baking-out process and the degassing under initial load can be carried out whilst the getter ion-pump takes over the task of the diffusion pump. Because of the small weight and dimensions it is now possible to transfer the tube together with the pump to the test equipment. Since the getter ion-pump is inexpensive, one can be fitted to each electron tube, and in certain circumstances it is possible for the pump to remain connected to the tube for life. Whereas it was formerly the normal practice to work with tubes on the pump, it is here rather a question of working with the pump on the tube.

Generally speaking, it is cheaper to use a getter ion pump in cases where long periods of evacuation would be necessary on the diffusion pump. It is also better to use a getter ion-pump when lower pressures than are normally required are needed for certain experiments.

We shall now consider three examples to demonstrate the application of our getter ion-pump.

In various experiments with a type of oscillator tube related to the magnetron⁸⁾, designed for high-power pulsed operation at decimetre wavelengths,

the object was to maintain a pressure of 10^{-6} torr during operation. The tubes had a volume of about 2 l and were all-metal, with a copper envelope and molybdenum electrodes. In the first place the tubes, to which an ion gauge and a getter ion-pump were connected, were evacuated on an oil-diffusion pump and degassed for 18 hours. The impregnated cathodes⁹⁾ of the tubes were heated up to about 1250 °C, and the cathodes of the getter ion-pump to about 900 °C at the end of the degassing period. Between the tube and the oil-diffusion pump two cold traps were mounted, one cooled with water and the other with liquid air. The pressure in the tubes, after seal-off from the diffusion pump, amounted to roughly 5×10^{-7} torr. When the tubes were afterwards put into operation, the pressure *without* the getter ion-pump in operation rose to 10^{-4} torr. With the getter ion-pump in operation, however, it proved possible, after an initial rise in pressure, to pump off the released gas rapidly and to maintain a pressure lower than 10^{-6} torr during the further operation of the tube.

As a second example we shall mention the application of the pump in various experiments on low-noise travelling-wave tubes¹⁰⁾. To keep the noise level low, the condition imposed on the vacuum in these tubes is that the pressure under load should not exceed 10^{-9} torr. The travelling-wave tubes, which deliver an RF power of a few microwatts at 4 Gc/s, have a glass envelope measuring 40 cm in length and 2 cm in diameter (approximate volume 0.15 l); the electron source is a barium-oxide coated cathode, and the helix is a molybdenum wire. The vacuum system, which consisted of the travelling-wave tube, an ion gauge and a getter ion-pump of the type described (fig. 6), was evacuated with diffusion-pump equipment described in this journal some time ago²⁾. During the last hours of the degassing period, which lasted 48 hours and during which the tube was baked out at a temperature of 400 to 450 °C, the electrodes of the ion gauge and the getter ion-pump were glowed. The oxide cathodes of the tubes were activated shortly before seal-off, while the whole system was still at a temperature of 200 °C. After seal-off the pressure amounted to 10^{-7} torr. The getter ion-pump reduced the pressure to 10^{-9} torr, and it was possible to maintain this pressure throughout operation of the tube.

In the examples given, the getter ion-pump was used to maintain the vacuum in the tube during operation.

⁸⁾ R. Levi, Philips tech. Rev. **19**, 186, 1957/58.

¹⁰⁾ Investigation, using the getter ion-pump, by A. Versnel. Philips Research Laboratories, Eindhoven.

⁸⁾ Investigated, using the getter ion-pump, by T. I. Sprenger, Philips Research Laboratories, Eindhoven.



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Fig. 6. Travelling-wave tube with getter ion-pump (above) and ion gauge (below) fused to one end. Top right, above the pump, can be seen the magnet for the getter ion-pump.

As a third example of the use that may in principle be made of a getter ion-pump, we mention in conclusion the evacuation of television picture tubes during manufacture ¹¹⁾. Tubes having screen diagonals of both 43 and 53 cm were evacuated by using solely a two-stage rotary backing-pump and a getter ion-pump. This method proved to be highly satisfactory. As it happens, however, its

application in mass production offers no special advantages over the usual method of pumping, which is already very economical.

Summary. For evacuating certain types of electron tubes, use can be made of a getter ion-pump. Because of its compactness, light weight and low cost, many special types of electron tubes can retain their own pump throughout their working life. This article describes a getter ion-pump using titanium as getter material. Ionization takes place as in a Penning vacuum gauge; the titanium itself sustains the Penning discharge. With its reserve of 50 mg of titanium the pump can remove a total of 2.5 torr.litre of CO. The maximum pumping speed is 62 l/sec. The lowest pressure achieved in small vacuum systems is roughly 10^{-10} torr.

¹¹⁾ These experiments were done partly by C. J. W. Panis and J. van der Waal in the Development Laboratory of the Electron Tubes Division, Eindhoven, and partly in the laboratory at Aachen.

ETCH PITS ON CADMIUM-SULPHIDE CRYSTALS

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The properties of cadmium sulphide for use in photoresistors, electroluminescent panels and other solid-state applications depend in large measure on the chemical impurities in the crystallites — a fact which has been the subject of much research. They also depend, however, on the physical imperfections of the crystal, and to investigate their influence, attempts are made to grow CdS crystals as nearly perfect as possible. In recent times various labora-

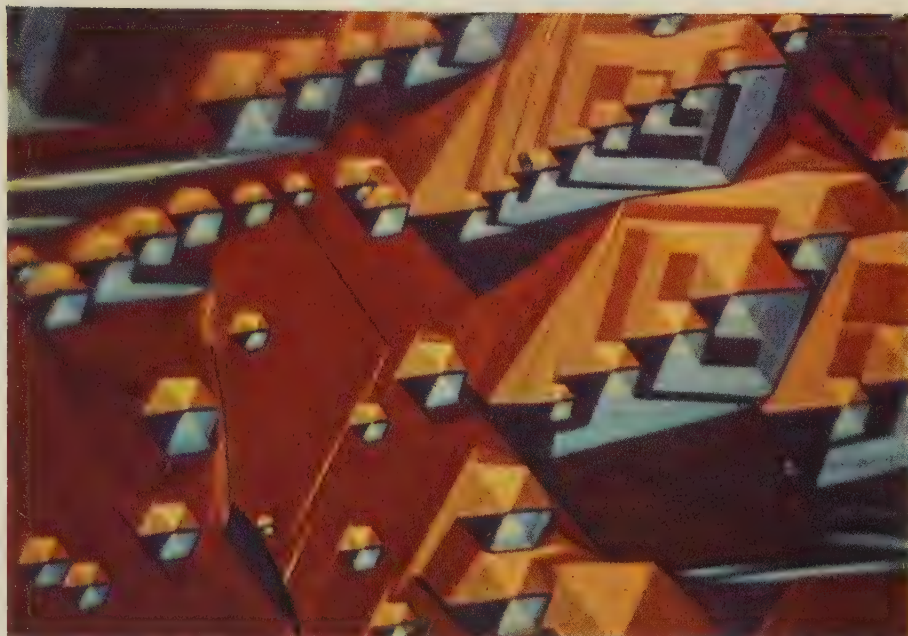
tories have succeeded in growing good single crystals of CdS having dimensions up to a few centimetres. CdS can occur in two crystal modifications — cubic and (polar) hexagonal. Under the present conditions of growth (sublimation at relatively high temperature) the hexagonal modification is obtained.

A crystal grown in this laboratory was etched on the basal plane in hydrochloric-acid vapour for about 1 minute (after wetting with water). Observed

under a microscope, using interference contrast ¹⁾, the etched surface shows colourful patterns of etch pits, two photographs of which are reproduced here (overall magnification about $\times 250$). The symmetry of the CdS crystal comes out beautifully in the hexagonal-pyramidal etch pits. It is not yet entirely certain how the formation of the steps visible in the pits should be interpreted.

Examination of the photographs reveals that many of the etch pits are disposed along straight lines, and that these lines are roughly parallel to one another, with only a few isolated pits between them. This may imply that the crystal possesses a high degree of perfection, for the following reasons.

For many crystalline substances a point-bottomed etch pit indicates the terminal point on the surface of a dislocation. A series of equidistant parallel dislocations running through the crystal lattice represents a "mosaic" boundary (sub-grain boundary) between two somewhat differently oriented crystalline blocks ²⁾. Assuming that this is the case here, the fact that the boundaries observed are few in number, and run more or less parallel, is in itself an indication that the structure of the crystal as a whole is good. Moreover, we can make an estimate of the difference in orientation between two adjacent mosaic blocks.



We consider here only the case of a symmetrical tilt boundary, made up of a row of parallel edge dislocations. (Similar considerations apply to other types of dislocation boundaries.) If b is the magnitude of the Burgers vector (the distance over which one part of the lattice is sheared with respect to the adjacent part), and d is the separation between successive dislocations (etch pits, assuming a 1:1 correspondence) along the grain boundary, then the angle between the two grain orientations is given by $\alpha \approx b/d$. We deduce from the photographs that d has the relatively large value of approximately 25 microns. Further, the grain boundaries are seen to lie roughly in the $\langle 10\bar{1}0 \rangle$ direction; the Burgers vector of the (edge) dislocations therein is then perpendicular to that direction, in other words in the $\langle \bar{1}210 \rangle$ direction, i.e. along the a -axis of the hexagonal structure. Assuming the magnitude of the Burgers vector to be equal to the lattice spacing in the $\langle \bar{1}210 \rangle$ direction, we calculate that $\alpha = b/d = 15 \times 10^{-6}$ radians $= 3''$. It would seem, therefore, that adjacent mosaic blocks in this crystal show extremely small differences of orientation, of the order of a few seconds of arc.

A. J. ELAND.

¹⁾ The technique used, which can make very small differences in height visible — often better than Zernike's phase-contrast method — was developed by Nomarski. (See G. Nomarski and A. R. Weill, *Rev. Metall.* **52**, 121, 1955.)

²⁾ See e.g. *Philips tech. Rev.* **15**, 246 and 286, 1953/54, for an introduction to the subject of dislocations, in which such concepts as the Burgers vector are explained; for polygonization to form mosaic boundaries, see p. 291.

SOLID-STATE RESEARCH AT LOW TEMPERATURES

III. THERMAL CONDUCTION IN INSULATORS; PARAMAGNETISM; DIELECTRIC LOSSES RELATED TO CHEMICAL LATTICE IMPERFECTIONS

by J. VOLGER.

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In this third and last article in the series on solid-state research at low temperatures ¹⁾ further examples are given of recent investigations. Those discussed here, however, are based on methods differing from those dealt with in the previous article, and are mainly concerned with research on insulators. Some of the low-temperature effects referred to here reveal the consequences of the fact that many processes proceed more slowly at low temperature.

Thermal conduction in insulators

The mechanism of thermal conduction in insulators closely resembles that of electrical conduction in metals. In the previous article ¹⁾ we saw that the electrical resistance of a metal, where the temperature is not too low, is primarily determined by the thermal vibrations of the crystal lattice, and at low temperature primarily by the scattering of electrons from lattice imperfections. In the transport of heat, analogous considerations apply to the lattice vibrations themselves, which both *constitute* the thermal energy and are the carriers or "transporters" of it (in insulators they are the *only* carriers). Whereas, in the case of metals, information on lattice imperfections can be derived from the behaviour of the electrical conductivity at low temperature, in the case of insulators — where the electrical method is obviously not applicable — similar information can be obtained by studying the thermal conductivity.

Before going into detail, we shall first say a few words about the Debye theory of the specific heat of solids, in which the underlying considerations on lattice vibrations are also fundamental to the problem of thermal conduction. This theory is based on the vibrational modes of the entire lattice. Thus where a lattice consists of N monatomic molecules, the number of vibrational modes (standing waves) $f(\nu)d\nu$ having a frequency between ν and $\nu + d\nu$ is given by:

$$f(\nu)d\nu = 9N\nu^2 d\nu / \nu_{\max}^3. \quad \text{(III, 1)}$$

Here, $\nu_{\max}$ is the maximum frequency of the thermal vibrations of the lattice; vibrations whose frequency is so high that the wavelength is smaller than the interatomic distance are irrelevant to a consideration

of the crystal as a whole ²⁾. The values of $\nu_{\max}$ are found on the basis of the premise of mechanics that the total number of independent modes of vibration of a system of N particles cannot be greater than $3N$, i.e.

$$\int_0^{\nu_{\max}} f(\nu)d\nu = 3N. \quad \text{(III, 2)}$$

The value of $\nu_{\max}$ that follows from this expression is found to correspond to a wavelength approximately equal to twice the interatomic spacing.

If, as in the first article for the case of molecules or atoms, we consider each vibrational mode as a quantized harmonic oscillator of zero-point energy $\frac{1}{2}h\nu$, then according to formula (I, 5) the average number of quanta of energy $h\nu$ that can be present per oscillator in thermal equilibrium is equal to $\{\exp(h\nu/kT) - 1\}^{-1}$. By analogy with the term photon for a light quantum — whose energy is also $h\nu$ — a quantum of elastic vibrational energy, or acoustic energy, is called a *phonon*. The number of phonons $g(\nu)d\nu$ found at a temperature T in the frequency range from ν to $\nu + d\nu$ is thus given by:

$$g(\nu)d\nu = \frac{f(\nu)d\nu}{\exp(h\nu/kT) - 1} = \frac{9N\nu^2/\nu_{\max}^3}{\exp(h\nu/kT) - 1} d\nu. \quad \text{(III, 3)}$$

It follows from this (cf. I, 6) that $g(\nu)$ is proportional to T at high temperatures — the total vibrational energy of the lattice is then proportional to T and the specific heat is constant — but not at low temperatures. This is represented graphically in *fig. 1*, where, omitting the factor $9N$, $g(\nu)$ is plotted in

¹⁾ J. Volger, Solid-state research at low temperatures, I. Introduction, Philips tech. Rev. 22, 190-195, 1960/61 (No. 6), and II. Electron conduction in metals and semiconductors, Philips tech. Rev. 22, 226-231, 1960/61 (No. 7).

²⁾ For the derivation of these formulae, see R. Kronig, Textbook of Physics, 2nd edn., Pergamon, London 1960. See also, for example, C. Kittel, Introduction to solid state physics, 2nd edn., Wiley, New York 1957, Chapter 6, and M. Born, Atomic physics, 6th edn., Blackie, London 1957, Chapter VIII, where the formulae are derived in somewhat different form.

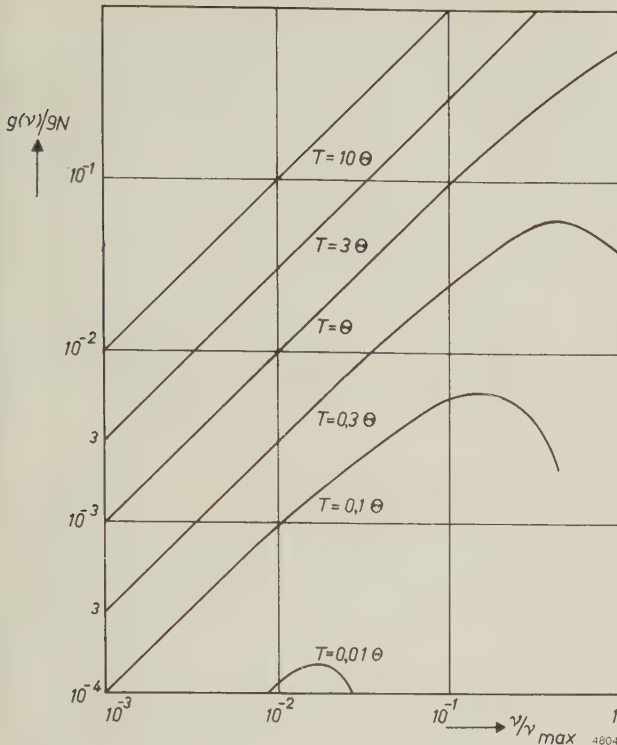


Fig. 1. The phonon spectrum in a solid. The number of phonons $g(\nu)$ of frequency ν (disregarding a factor $9N$) is plotted versus frequency for various temperatures. The temperature T is expressed in the figure as a fraction of the Debye temperature Θ , the frequency as a fraction of the maximum frequency $\nu_{\max}$

log-log coordinates against frequency (as a fraction of $\nu_{\max}$), for various values of the ratio T/Θ (Θ is the Debye temperature, given by $h\nu_{\max}/k$). It can be seen that at high temperature the curves are straight lines which displace vertically by one decade for every increase in temperature by a factor of 10. For $T \ll \Theta$, this remains valid only at low frequencies; quanta with frequencies in the region of $\nu_{\max}$ become fewer in number as the temperature decreases and finally are virtually non-existent. The maximum of the curve lies approximately at the frequency for which $\nu/\nu_{\max} = 1.6 T/\Theta$. This behaviour affects the variation at low temperatures of both the specific heat and the thermal conductivity.

According to Debye's theory the total energy E is given by:

$$E = \int_0^{\nu_{\max}} g(\nu) h\nu d\nu, \quad \dots \quad (\text{III}, 4)$$

from which we find $E = 3NkT$ for the high-temperature region, where the specific heat thus has the required constant value $3Nk$, and at low temperatures:

$$E = \frac{\pi^4}{5} \left(\frac{kT}{h\nu_{\max}} \right)^3 3NkT. \quad \dots \quad (\text{III}, 5)$$

It follows from this theory, then, that the specific heat at low temperature is proportional to T^3 , which agrees very well with the variation found by experiment.

For a solid, in which the heat transport can be represented by the movement of phonons, the formula for the coefficient of thermal conductivity κ , that is the ratio between the quantity of heat flowing per unit time through unit area and the temperature gradient, is arrived at by a method similar to that applied in the case of a gas. According to kinetic theory, the thermal conductivity of a gas is given by:

$$\kappa = \gamma C v l, \quad \dots \quad (\text{III}, 6)$$

where C is the heat capacity per unit volume, v the arithmetic mean velocity of the molecules, l their mean free path and γ a constant. The thermal conductivity of solids is expressed by exactly the same formula, except that phonons should be read instead of molecules in the definition of v and l . The velocity v , then, is now the velocity of sound in the material concerned. The value of γ is of the order of magnitude of unity.

Before considering how κ varies with T in the low-temperature region, it should be recalled that the phonons fulfil two functions. In the first place they *constitute* the thermal energy, and in the second place they are responsible for the transport of the thermal energy. In a crystal where the phonons were able to move quite freely, the heat transport would thus be comparable with the transmission of energy in free space by electromagnetic radiation. In a real crystal this is not the case. Due to the scattering of the phonons for one reason or another, the heat transport takes on the character of diffusion³⁾.

We shall now discuss the behaviour of κ with decreasing temperature by reference to a curve⁴⁾ measured on bismuth telluride (Bi_2Te_3), represented in fig. 2. In this figure $1/\kappa$ is plotted versus T . The

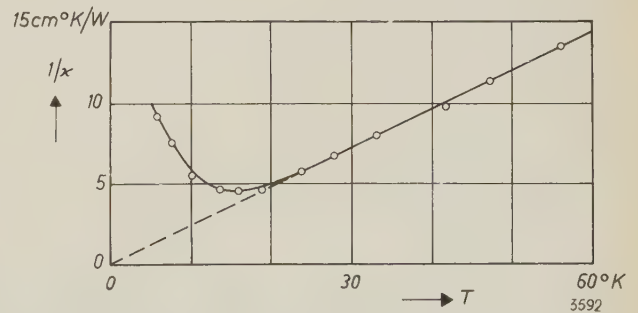


Fig. 2. The reciprocal of the coefficient of thermal conductivity κ of bismuth telluride as a function of temperature.

- ³⁾ Macroscopic evidence of this is that the heat flow is proportional to the temperature gradient. If there were no scattering, the heat flow through a bar would be proportional to the temperature difference between its ends, irrespective of its length.
- ⁴⁾ Measurements by G. Bosch of this laboratory.

graph shows clearly that above 20 °K the curve is virtually a straight line passing through the origin, indicating that in this region κ is proportional to $1/T$. The form of the curve may be explained as follows. If the temperature is appreciably higher than the Debye temperature Θ , then, as we have seen (eq. III, 3 *et seq.* and fig. 1), the value of the phonon distribution function $g(\nu)$ is proportional to T . Now, in sufficiently pure crystals the scattering of phonons in this temperature range is primarily due to their mutual "collisions". The probability of such a collision is proportional to the number of phonons, and hence proportional to T . The mean free path l is therefore inversely proportional to T , and the same holds for κ , since the other quantities in formula (III, 6) are constant in this temperature range.

When T approaches Θ (at 150 °K in the case of Bi_2Te_3), the number of phonons decreases more than proportionally with T , and the same applies to the probability of their being scattered, but here the specific heat also begins to decrease. At first, these effects roughly compensate one another. This partly explains why the proportionality between κ and $1/T$ continues to exist quite a long way below the Debye temperature.

If the collisions between the phonons were the only cause of the scattering, then at very low temperatures ($T \ll \Theta$) the thermal conductivity would finally increase very steeply with falling temperature. It can be seen from fig. 2 that the reverse is the case. Since $g(\nu)$ is very small in this temperature range, and so is the probability of collisions between the phonons, the scattering from lattice imperfections begins to play a relatively important part here. A calculation shows that, as a consequence of this, the mean free path l of the phonons varies only to a slight extent with temperature for $T < \Theta$. The specific heat however, at $T < \Theta$, varies with T^3 , so that with decreasing temperature κ must indeed undergo a sharp decrease.

If the lattice contained no defects whatever, the mean free path with falling temperature would finally be determined solely by the dimensions of the material, and would thus be independent of temperature. The thermal conduction in that case would be comparable with a Knudsen flow, i.e. the flow of a gas which is so rarefied that the number of molecular collisions is negligible compared with the number of collisions with the wall of the tube through which the gas is flowing, or it might be compared with the movement of light quanta in an internally reflecting tube (see ³). An effect of this kind has in fact been found experimentally, for example on diamond and on some alkali halides ⁵).

⁵) W. J. de Haas and T. Biermasz, *Physica* **5**, 47, 320 and 619, 1938. A theoretical treatment is given by H. B. G. Casimir in *Physica* **5**, 495, 1938. See also R. Peierls, *Ann. Physik* **2**, 5, 1055, 1929.

Summarizing, it can be said that the variation of thermal conductivity as a function of temperature provides important information on the scattering of phonons by lattice imperfections; indeed, the study of the thermal conduction of insulators is no less important as a research tool than the study of electron conduction in metals. The way in which the phonon spectrum varies with temperature, has, of course, an equally important bearing on the variation of other processes which take place under the influence of lattice vibrations. A notable example of this will be discussed in the next section.

Paramagnetism

From formula (I, 8) for the magnetic moment of paramagnetic substances we were able to deduce that from the point of view of the magnetization, we may speak of a "low" temperature where $kT \ll \mu_B H$. In this temperature range the magnetization is no longer proportional to H , as it is in the case of normal field strengths at room temperature, but saturation occurs (fig. 2 in I).

We have already mentioned that the rate at which some processes take place in the solid state is slowed down when the temperature falls. We shall now examine this effect more closely for the case of paramagnetism. The application of an external magnetic field does not instantaneously produce the magnetization $\bar{\mu}$ given by (I, 8); some time elapses before the new state of equilibrium is reached. If this orientation process, called paramagnetic relaxation, is an exponential function of time, it can be described by a characteristic time τ , the relaxation time. The cause of the relaxation phenomenon is of a very general nature. A paramagnetic ion can orient itself in a magnetic field only if it is able to exchange energy with its surroundings, just as, for example, a pendulum pulled out of equilibrium returns finally to its vertical position under the action of gravity only because its movement is damped. If the ion is unable to give up energy, it will continue to describe a precessional motion without any decrease in the angle between the directions of $\bar{\mu}$ and H , so that the component of $\bar{\mu}$ in the H direction does not change. In such a case the pendulum in our example would go on swinging with undiminished amplitude. It may thus be concluded that the magnitude of the relaxation time is determined by the extent to which energy can be exchanged.

Since τ is generally small, the inertia of the orientation process is not directly evident when a constant magnetic field is applied, but it is apparent when the

magnetizing force is periodically varied. As long as the frequency is small compared with the reciprocal of the relaxation time, no effect is observed. When the frequency is raised, however, the susceptibility in the frequency range around $1/\tau$ begins to decrease. A phase shift also occurs between H and $\bar{\mu}$. Both effects indicate that the orientation of the paramagnetic ions can no longer properly follow the variations of the magnetic field.

In order to give a qualitative explanation for the marked increase which τ must undergo with falling temperature, we shall return for a moment to the case touched on in the first article, where the spin can assume only two values, namely parallel and anti-parallel to the magnetic field. It was mentioned there that the energy difference between the two states is $2\mu_B H$. In a transition, then, the ion must either absorb or give up this amount of energy, depending on the direction of the transition. This it can do, via the lattice vibrations, in two ways. In the first place, a phonon of energy $2\mu_B H$ may be absorbed or emitted, and in the second place a phonon may be non-elastically scattered by the ion, and thereby give up or absorb the energy $2\mu_B H$. The latter process is directly comparable with the Raman effect, familiar in optics. Both processes have a greater probability of occurring the more phonons are available. As regards the Raman process, this is immediately understandable; as regards the spin-phonon process it is also understandable in so far as it concerns the *absorption* of a phonon. Quantum mechanics shows, however, that the chance of the *emission* of a phonon is also greater the greater the number of phonons. As in the case of absorption, they must likewise have the energy $2\mu_B H$ in order to be effective.

In the foregoing section (see fig. 1) we have already seen that the phonons do indeed decrease sharply in number as the temperature falls. It is therefore clear that the relaxation process will be slower, and the relaxation time τ longer, the lower is the temperature. Fig. 3 shows the variation with temperature of the relaxation time for gadolinium sulphate and for a chromium-alum.

Apart from the spin-lattice relaxation just discussed, a direct exchange of energy is possible between the ions mutually. The extent to which this process can take place is determined by the strength of the ionic interaction and therefore depends on the concentration of the paramagnetic ions. This interaction may thus be attenuated by "diluting" a salt with a certain quantity of non-magnetic ions. In normal, undiluted paramagnetic substances the relaxation time τ_{s-s} corresponding to this spin-spin relaxation is of the order of magnitude of only 10^{-10} sec. In diluted salts, τ_{s-s} is accordingly somewhat

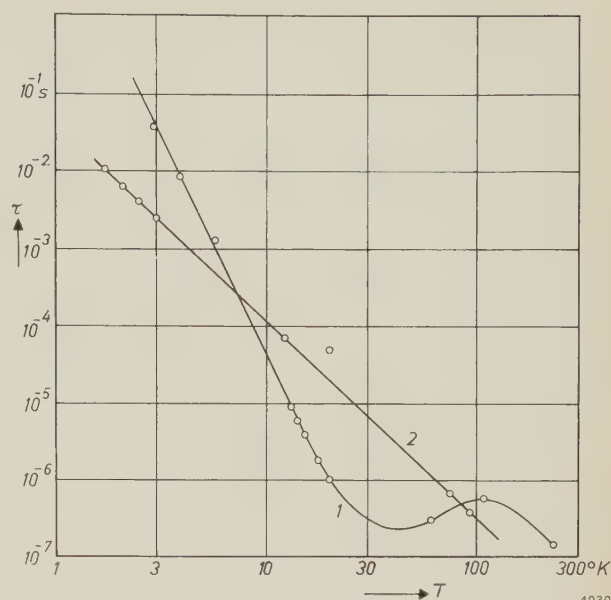


Fig. 3. The characteristic time τ of a paramagnetic relaxation increases steeply as the temperature falls. The figure gives a log-log plot of τ (in seconds) versus temperature, as measured on two substances:

- 1 : $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$; measured at 2.6×10^5 A/m;
2 : $\text{CrK}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$; measured at 3.2×10^5 A/m.

longer. The speed of this relaxation phenomenon is not affected by temperature⁶⁾.

Paramagnetic resonance

Temperature is a factor of importance in various respects in solid-state research using the method of paramagnetic resonance, which has already been described at some length in this journal⁷⁾. In this method the degeneracy of the ground state of the paramagnetic ions is removed by the application of a magnetic field, the energy level thereby being split into a number of sub-levels the distance between which, ΔE , is proportional as a first approximation to H . Transitions between neighbouring sub-levels can be brought about by irradiating the substance with electromagnetic waves whose frequency ν_r is such that $h\nu_r = \Delta E$. Where most of the ions occupy the lower of two such levels — which is the normal situation — the result is a net absorption; in the contrary case a net emission of radiation having the frequency ν_r occurs.

It is necessary to speak of *net* absorption and *net* emission because in each case both emission and absorption take place;

⁶⁾ The existence of a temperature-independent relaxation phenomenon side by side with the spin-lattice relaxation, which does vary with temperature, was predicted on theoretical grounds by J. Waller in 1932 (Z. Phys. **79**, 370, 1932). The existence of paramagnetic relaxation effects was first demonstrated experimentally by C. J. Gorter (Physica **3**, 503, 1936).

⁷⁾ J. S. van Wieringen, Paramagnetic resonance, Philips tech. Rev. **19**, 301-313, 1957/58.

in the first case the number of absorbed quanta exceeds the number of emitted quanta, in the other case the opposite applies. Both numbers are proportional to the population of the two levels and to the number of incident quanta. The fact that the emission is proportional here to the intensity of the incident radiation field is an indication that it is *stimulated* emission. Unlike the situation in optical spectroscopy, spontaneous emission is negligible here (cf. reference 7)).

The temperature has a threefold influence on the observed phenomena. In the first place it determines the degree to which one can discriminate between the details of the absorption spectrum; secondly it determines the ratio between the intensity of the incident and the absorbed radiation — i.e. the sensitivity —, and thirdly it governs the maximum permissible intensity of the incident radiation.

As regards the discrimination between details of the resonance spectra, it should be recalled that the energy levels into which the ground state is split are often themselves split into a number of very closely spaced sub-levels. It is precisely the latter splitting that provides the most important information on the substance under investigation. These fine details obviously cannot be observed if the width of the lines in the resonance spectrum is greater than the distance between adjacent sub-levels. Since this width is related to the rate of the relaxation process (the slower the relaxation process the smaller the line width), it is possible in appropriate cases, e.g. in investigations on substances containing Ti^{3+} , Fe^{2+} or Co^{2+} , to achieve a sufficiently small line width by reducing the temperature. A good example is shown in fig. 4.

With regard to the temperature-dependence of the sensitivity it should be recalled that the net absorption is greater the greater the excess population of the lower of the two levels between which the transitions take place. The ratio between the populations of the two levels is proportional to the Boltzmann factor $\exp(-\Delta E/kT)$, and therefore the absorption can be increased by lowering T . Since ΔE is of the order of magnitude of $k \times 1^\circ\text{K}$, this would produce in all cases an appreciable gain if it were not for the fact that the temperature reduction also gives rise to an unfavourable effect — the third temperature effect mentioned above.

In a resonance experiment the population of the levels is not exactly that given by the Boltzmann distribution, the reason being that the radiation field causes transitions to the upper level, which it thus tries to “pump full”. At normal temperatures, where the relaxation time is short (e.g. 10^{-7} sec) and where consequently the upper level is soon

“emptied” again, the disparity is slight and the sensitivity is not seriously affected. When the sample is cooled, however, the relaxation time increases, as we have seen, and in some cases a situation may be reached where the population of the upper level is equal to that of the lower level (saturation), so that there is virtually no net absorption. In that case, to avoid saturation, one would have to work with a considerably weaker incident radiation. The choice of the temperature and the intensity of the incident radiation is therefore always a compromise, to be decided from one case to another.

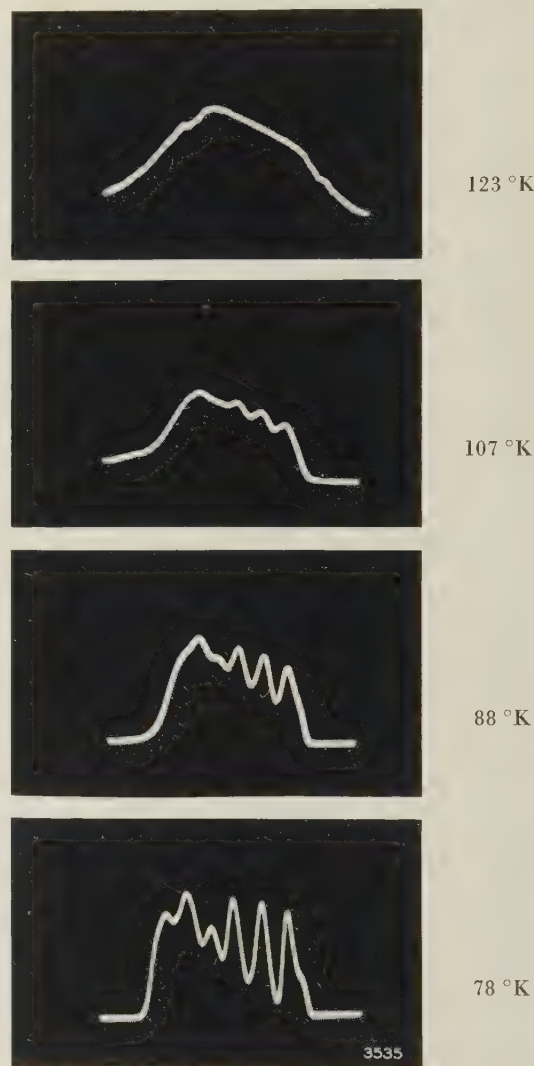


Fig. 4. Spectral line with hyperfine structure, from the paramagnetic resonance spectrum of α -quartz irradiated with X-rays. This irradiation produces colour centres in the quartz near aluminium impurity atoms. The recordings were made at four different temperatures. It can be seen that each of the fine-structure lines becomes narrower — and therefore higher, since their integrated area remains unchanged — the lower is the temperature. At the highest temperature their width is so large relative to their separation that they are merged into each other. The frequency of the electromagnetic field was 9.7 Gc/s ($\lambda = 3.1$ cm). The orientation of the permanent magnetic field was parallel to that of the c axis of the quartz crystal. (Measurements by J. S. van Wieringen of the Philips Research Laboratories, Eindhoven.)

Solid-state amplifiers for microwaves

We shall now consider, again from the point of view of temperature, the microwave amplifiers which have been given the name of maser (*Microwave Amplification by Stimulated Emission of Radiation*). We shall be concerned only with the so-called three-level solid-state maser⁸). It will be shown that the physical principles underlying the temperature effects here encountered are largely the same as those just discussed under the heading of paramagnetic resonance.

The operation of the maser depends on the fact, already touched upon, that when a paramagnetic substance is exposed to radiation of a frequency corresponding to the energy difference between two levels, the result is a net emission — and hence amplification of the incident signal — if the population of the upper level is greater than that of the lower level. In that case the transitions to the lower level exceed the transitions to the upper one.

The reason for this is evident when it is realized that, like the probability of absorption — i.e. of an upward transition — the probability of emission is also proportional to the relative populations of the energy levels and to the intensity of the incident radiation; the respective proportionality factors are the same for absorption and emission. The emission is almost entirely stimulated emission; the probability of spontaneous emission, which of course depends on the population of the upper level but is not affected by the incident radiation, is negligible or at least of minor significance.

The *inversion* of the energy-level population, on which the operation of the maser depends, can of course only be brought about by some external intervention; in a substance in thermal equilibrium the lower level always has the greater population.

In the Bloembergen solid-state maser a paramagnetic salt whose ground state, in a magnetic field, splits into at least three sub-levels, is made emissive by irradiating it with microwave energy corresponding to the transition between the lowest and highest of the three levels (1 and 3, respectively, in *fig. 5*). The intensity of the microwave radiation is chosen high enough to produce saturation, i.e. such that the population of 3 is equal to that of 1. Evidently, this is only feasible in practice provided the relaxation time τ_{31} for the return to the lowest level is not unduly short. This situation can be improved by reducing the temperature of the substance, thereby reducing the required "pumping power" to an attainable value. Here, then, the effect which was

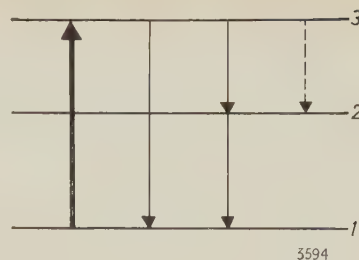


Fig. 5. Illustrating the operation of a solid-state maser, after Bloembergen. A paramagnetic salt is used whose ground state in a magnetic field splits into at least three levels.

a disturbing effect in the resonance measurements is turned to good use.

Now an ion, in addition to returning directly from level 3 to level 1, may also return by first moving to level 2, thereby emitting one or more phonons of intermediate energy (thin lines in *fig. 5*). It can be calculated that the population N_2 of level 2 in the state of equilibrium is determined not only by the temperature but also by the energy differences E_{12} and E_{23} and the relaxation times τ_{32} and τ_{21} of these transitions. It further appears⁸) that level 2 will be less populated than level 3, and that consequently inversion will occur where:

$$\frac{E_{12}}{\tau_{21}} > \frac{E_{23}}{\tau_{32}} \quad \dots \quad (\text{III}, 7)$$

If $E_{12} \approx E_{23}$ (the two energies should not be identical!) this roughly amounts to the condition $\tau_{32} > \tau_{21}$. In other words, level 2 must be "emptied" faster than level 3. The maximum positive difference ($N_3 - N_2$) between the populations is given by:

$$(N_3 - N_2)_{\max} = N \times \frac{1 - \exp(-E_{12}/kT)}{2 + \exp(-E_{12}/kT)} \approx \frac{N E_{12}}{3 kT} \quad \dots \quad (\text{III}, 8)$$

where $N = N_1 + N_2 + N_3$. It can be seen that this difference is greater the lower the temperature of the salt. For this reason, the masers we are now discussing have hitherto been designed to operate at the temperature of liquid helium.

When electromagnetic radiation of frequency ν_{23} ($= E_{23}/h$) is incident on a paramagnetic salt whose energy levels and relaxation times are such as to satisfy (III, 7) and which is supplied with a sufficient pumping power, this radiation — owing to the inversion of the populations of 2 and 3 — induces the emission of stronger radiation of the same frequency (dashed line in *fig. 5*). The salt then functions as a maser. (If τ_{32} is smaller than τ_{21} , the population of 1 and 2 is inverted, and the result is a maser which amplifies radiation of the frequency ν_{12} .)

Of the three categories of amplifiers now known

⁸) The idea underlying this type of maser is due to N. Bloembergen (Phys. Rev. **104**, 324, 1956).

— namely radio-valve (or transistor) amplifiers, parametric amplifiers, and masers — the masers have the lowest noise figures. For this reason, in spite of their obvious drawbacks — cooling with liquid helium and amplification only in an extremely narrow waveband — masers nevertheless have interesting technical possibilities.

The noise of a maser is mainly due to the spontaneous transitions from the upper to the lower level, which, unlike the stimulated transitions, are of course not correlated with the incident radiation. This noise contribution may be expressed theoretically as the thermal noise of a negative resistance at a negative temperature⁹⁾. This negative temperature is that which would have to be substituted in the Boltzmann factor in order to describe the inverted population of the energy levels. Since the absolute value of this temperature is of the same order of magnitude as the true temperature, it is understandable from the point of view of noise reduction that the maser should be operated at a low temperature. This also reduces, incidentally, the thermal noise of the resonant cavity in which the crystal is irradiated.

Cooling by adiabatic demagnetization

Although the subject does not, strictly speaking, come within the scope of the present article, it will be useful at the end of this section on paramagnetism to touch briefly on the celebrated method of cooling based on the adiabatic demagnetization of a paramagnetic salt¹⁰⁾. A "pellet" of a suitable paramagnetic material, subjected to a strong magnetic field, is in thermal contact on one side with the sample to be cooled, and on the other with a bath of boiling helium. Once thermal equilibrium is established, the thermal contact with the helium bath is broken, after which the magnetic field is switched off. The temperature of the pellet then falls. Ingenious application of this principle has made it possible to reach a temperature of 0.001 °K.

The physical background of the method may be briefly explained with reference to *fig. 6*, which shows schematically the variation of the entropy S as a function of temperature, both in the presence and the absence of a magnetic field. Since the magnetized state of the salt represents a state of greater order than the non-magnetized state, the entropy at any given temperature is greater in the absence of a magnetic field. Upon adiabatic (isentropic) demagnetization — stage 2 in *fig. 6* — the temperature of the paramagnetic material therefore drops.

A very pronounced effect can be produced with salts whose ground state, when no external magnetic field is applied, may be regarded to a first approximation as completely degenerate. This means that the splitting of the ground state caused by the internal crystalline field, which therefore remains after the external field is removed, gives rise to only very small energy

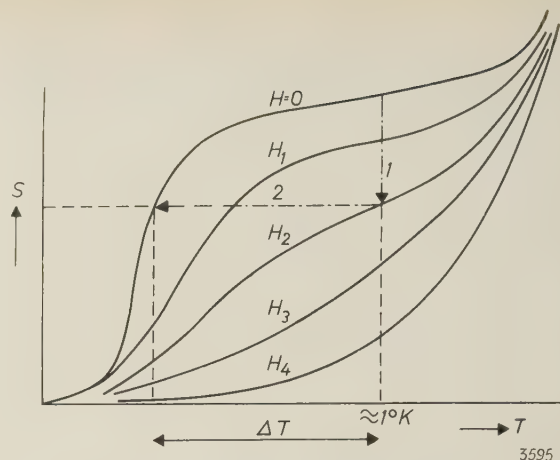


Fig. 6. The variation of the entropy S of a paramagnetic salt as a function of temperature T in the region between 0 and 1 °K, in the presence of magnetic fields of varying strengths. A temperature drop ΔT can be brought about by cooling the salt to about 1 °K, applying the magnetic field H_2 (stage 1), breaking the thermal contact with the environment, and finally switching off the magnetic field (stage 2).

differences between the sub-levels. The greater this so-called zero-field splitting, the greater is the order prevailing in the non-magnetized crystal, and hence the smaller is the entropy difference between the magnetized and the normal state; the temperature reduction that can be achieved will therefore likewise be smaller.

Dielectric losses in relation to chemical lattice imperfections

In the theory of dielectrics the magnitude of the dielectric constant can be related to various mechanisms of polarization. For example, there is electronic polarizability, which arises from the displacement of the electrons relative to the nucleus; there is ionic polarizability, due to the relative displacement of ions of opposite sign in the crystal lattice; and there is orientational polarizability in substances containing molecules which have a permanent dipole moment. Since solid-state research in recent decades has evolved primarily into an investigation of lattice imperfections, we shall confine ourselves here to a few examples of dielectric phenomena related to lattice imperfections¹¹⁾. In particular we shall be concerned with cases where an imperfection contains an electric dipole of moment p , the orientation of which in an external electric field is subject to a certain inertia; the manner of orientation can be described in our examples by a single characteristic time, the relaxation time τ .

As in the case of paramagnetism, discussed in the previous section, where the inertia of the relaxation process leads to a lower susceptibility when the frequency of the field variations is greater than $1/\tau$,

⁹⁾ R. V. Pound, *Ann. of Physics* **1**, 24, 1957.

¹⁰⁾ This subject is dealt with in detail by C. G. B. Garret, *Magnetic cooling*, Harvard, Cambridge, 1954, and by D. de Klerk in an article in *Handbuch der Physik*, Part XV, p. 38-209, Springer, Berlin 1956.

¹¹⁾ A survey of this field is given by J. Volger in *Dielectric properties of solids in relation to imperfections*, *Progress in Semiconductors* **4**, 205-236, 1960.

here too we find that the dynamic value of the dielectric constant decreases when the frequency of the applied electric field increases to values higher than $1/\tau$ ¹²). Let ϵ_m be the relative dielectric constant of the unperturbed lattice — which, for convenience, we shall regard as constant — then the value ϵ found at a certain angular frequency ω is given by:

$$\epsilon = \epsilon_m + \frac{\Delta\epsilon}{1 + \omega^2\tau^2}. \quad \dots \quad (\text{III, 9})$$

The value of the quantity $\Delta\epsilon$ follows from:

$$\Delta\epsilon = Nf \frac{4\pi p^2}{3kT}. \quad \dots \quad (\text{III, 10})$$

Here N is the number of dipoles per unit volume, and f is a factor defining the effect of the internal (electrical) crystalline field (the dipoles are not affected solely by the applied external field, but by the resultant of this field and the internal field). Provided the concentration N of the dipoles is not too high, f is given by:

$$f = (\epsilon_m + 2)^2/9. \quad \dots \quad (\text{III, 11})$$

Both the relaxation time τ and the quantity Np^2 can be determined directly. The latter quantity is found by measuring ϵ at high and low frequency; with the aid of (III, 9) the value of $\Delta\epsilon$ can then be found, and substituted in (III, 10). The relaxation time can be measured in principle by ascertaining the frequency range in which ϵ changes in value; this is not very accurate, however, because the change in ϵ is only a small fraction of its value.

A better method of determining τ is to ascertain the variation with frequency of the loss angle δ , i.e. the phase angle between the applied alternating field and the dielectric displacement. This angle can be found by measuring the dielectric losses, which are proportional to $\tan \delta$. The variation of $\tan \delta$ with frequency is expressed by the formula:

$$\tan \delta \approx \frac{\Delta\epsilon}{\epsilon_m} \frac{\omega\tau}{1 + \omega^2\tau^2}. \quad \dots \quad (\text{III, 12})$$

It can be inferred from this expression that $\tan \delta$ depends strongly on frequency and shows a peak at $\omega = 1/\tau$.

In many cases the variation of τ with frequency is given by an expression of the form:

$$\frac{1}{\tau} = \frac{1}{\tau_0} \exp(-Q/kT). \quad \dots \quad (\text{III, 13})$$

¹²) An extensive treatment of the theory of dielectric phenomena, giving the derivation of the formulae mentioned in this section, will be found in: H. Fröhlich, *Theory of dielectrics*, Oxford University Press, London 1950.

The energy Q in this expression is an "activation energy", representing the height of the energy barrier which an ion or an electron must exceed to arrive at the new state. It is plausible that the formula expressing the probability of this barrier being exceeded will contain a Boltzmann factor, and so therefore will the formula for τ . The nature of this energy barrier will be discussed in more detail presently. In some cases an ion can reach a new state as a result of a wave-mechanical "tunnelling" effect, or because the energy barrier which keeps it entrapped disappears for a moment owing to a favourable combination of the movement of other ions. In these cases too, an expression of the form (III, 13) is often applicable. This formula brings out clearly the important bearing which the temperature has on the study of dielectric phenomena. Especially if Q is small, the relaxation time at high temperature is so short that its reciprocal lies outside the accessible frequency range. Only by working at low temperatures is it possible to observe losses corresponding to such a mechanism.

It should be added that the analogy with paramagnetic relaxation — where formulae (III, 9), (III, 10) and (III, 12) also apply, *mutatis mutandis* — ceases to be valid as regards the nature of the relaxation mechanism. The manner in which τ varies with T is often quite different, even though τ may be large at low temperatures and small at high temperatures.

Dielectric losses in quartz due to local deformation of the crystal lattice

Dielectric losses attributable to a polarization mechanism of low activation energy, which are therefore observable only at low temperature, are found (for example) in quartz. Fig. 7 shows the result of measurements of $\tan \delta$ as a function of T — and thus indirectly as a function of τ — on three different samples at a frequency of 32 kc/s. The curves are seen to exhibit a number of peaks. The temperatures at which these peaks occur — in so far as they are present in a particular curve — are the same for all three curves shown. Spectrochemical analysis of the crystals revealed the impurities which they contained, and on this basis an attempt was made to correlate each peak with the presence of a particular kind of foreign atom. On the assumption that formulae (III, 10), (III, 12) and (III, 13) are applicable here, one can calculate for the temperature T at which a peak occurs that:

$$kT_m = -\frac{Q}{\ln \omega\tau_0} \left(1 - \frac{1}{(\ln \omega\tau_0)^2} + \dots \right). \quad (\text{III, 14})$$

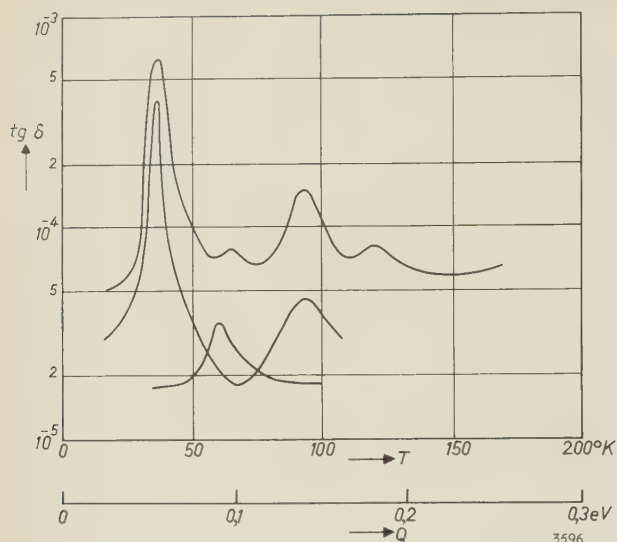


Fig. 7. The variation of $\tan \delta$ with temperature T for three quartz samples in an alternating electrical field of 32 kc/s. The peaks observed are attributed to polarization mechanisms connected with the presence of impurities. The values of the activation energy Q corresponding to each peak can be read from the lower abscissa scale.

Since τ_0 proved to have the value 10^{-13} sec for all peaks in all three samples, it was possible to draw under the temperature scale in fig. 7 a linear energy scale, applicable to all three samples. It can be seen that the Q values are of the order of 0.1 eV. This is indeed very small; the Q value for the displacement of an ion in the crystal lattice — that is to say a displacement from one lattice site to another — corresponds to a value roughly 10 times as high.

The mechanism of the polarization processes responsible for these losses is assumed to be roughly as follows. It is assumed that in "wide-mesh" crystal lattices, as found in quartz and glass, the potential wells which correspond to the lattice sites, have a "sub-structure", each consisting of a small group of separate minima. These sub-minima are thus separated from one another by tiny potential barriers whose height is much less than that of the maxima between the lattice sites (fig. 8). Even at temperatures so low that an ion cannot possibly



Fig. 8. The occurrence of dielectric losses at such a low temperature that an ion cannot possibly move from one lattice site to another is explained by assuming that, in "wide-mesh" lattices (quartz, glass, etc.), the lattice sites do not always correspond to a simple minimum of the potential energy E , but to a group of sub-minima. The drawing illustrates a cross-section of the potential variation in such a lattice site in which two sub-minima occur.

move from one lattice site to another, the ions are still capable of a slight displacement, i.e. from one sub-minimum to another. The small value of Q for these processes corresponds to the low height of the potential barriers separating the sub-minima.

The multiple minima are assumed to be present mainly in the immediate proximity of foreign atoms. The details of the structure of chemical lattice imperfections are unfortunately not yet sufficiently known to be able to say with certainty in exactly what way the above-mentioned polarization processes take place in an actual crystal.

Dielectric losses in oxidic semiconductors

Another remarkable polarization mechanism, which is likewise only observable at low temperature, is found in oxidic semiconductors. We shall discuss its nature with reference to the case of nickel oxide containing trace quantities of lithium.

In the nickel-oxide lattice the Ni ions are divalent and positive. If one of the Ni ions is replaced by an Li ion, the local electric neutrality is disturbed, since Li ions are monovalent, and there is no evidence of the existence of divalent Li ions. As the ions of the iron group do possess different valencies, an obvious supposition is that the neutrality might be restored by the presence of a trivalent Ni ion in the immediate vicinity of the Li^{1+} ion (fig. 9).

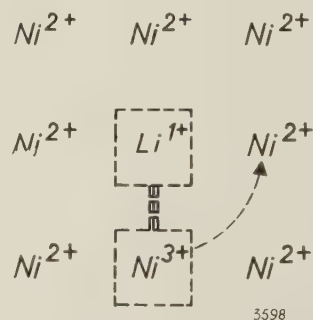


Fig. 9. The electric neutrality of a nickel-oxide crystal containing Li^{1+} ions is maintained by one of the surrounding Ni ions becoming trivalent. Due to the transference of an electron, the trivalence may move to any of the Ni ions that are immediate neighbours of the Li ion. This mechanism is proposed to explain the dielectric losses found at low temperature in impure or non-stoichiometric oxidic semiconductors.

At a temperature so low that all electrons are bound to the donors, the material thus being an insulator, dielectric losses are found to occur.

It is assumed that the polarization, i.e. the alignment of the dipole formed by the Li^{1+} ion (which is negatively charged relative to the surrounding Ni^{2+} ions) and the Ni^{3+} ion (which has a relative positive charge) is brought about by an electron transferring from one of the Ni^{2+} ions to the Ni^{3+} ion, apparently causing the latter to change place.

The Ni ions likely to be involved are only those surrounding the Li^{1+} ion. This is somewhat analogous to the loss mechanism discovered by Breckenridge which occurs in such types of point defect. An example, discussed in this journal some time ago¹³⁾, is the case of a substitutional Ca^{2+} atom in the NaCl lattice, where the electric neutrality is restored by the presence of a vacancy on an Na site. This vacancy can occupy any of the Na sites around the Ca^{2+} atoms, but cannot get away at moderate temperatures.

The same phenomenon observed on nickel oxide has been found in $\alpha\text{-Fe}_2\text{O}_3$, where Ti^{4+} ions were substituted for a number of Fe^{3+} ions, and also in non-stoichiometric $\alpha\text{-Fe}_2\text{O}_3$. In the first case the electric neutrality is assumed to be restored as a result of one of the Fe ions around the Ti^{4+} becoming divalent, and thus possessing an extra electron. Fig. 10 shows the result of measurements on non-

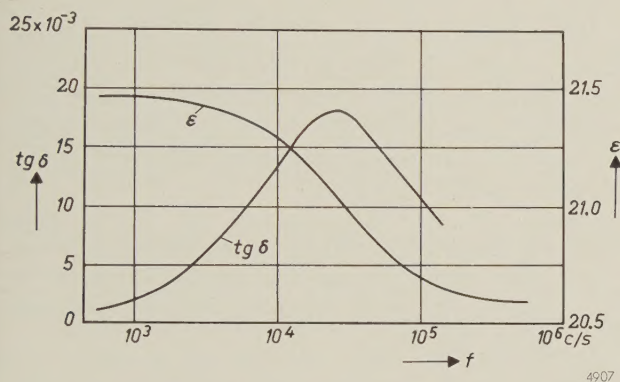


Fig. 10. Curves of the dielectric constant ϵ and of the loss $\tan \delta$ as functions of the frequency f , for a non-stoichiometric crystal of $\alpha\text{-Fe}_2\text{O}_3$ at a temperature of 20 °K.

stoichiometric $\alpha\text{-Fe}_2\text{O}_3$. The relaxation time τ was governed here by the extremely small activation energy of 5×10^{-3} eV and a relaxation time τ_0 of 2×10^{-7} sec.

A strong argument for the assumption that the polarization mechanism is bound up with the presence of foreign, or at least extra, atoms, is the fact that the losses rise and fall with their concentration. In a pure state the substances do not show the effect at all.

To conclude this series of articles, a few comments will not be out of place on the technological significance of cryogenic solid-state research. Its importance, as in the case of all more or less fundamental research, may be said to be both direct and indirect. As regards the latter, it should be noted that the study of each of the effects discussed — including those that have no direct technical importance — forms part of a wider programme of research, any part of which might yield the essential factor for obtaining new insight that can be turned to practical ends. To give only one instance, the study of paramagnetic resonance and dielectric losses has increased our knowledge of lattice imperfections, and the greater insight thereby gained into the discolouration shown by certain types of glass, subjected to intense irradiation, has proved to be extremely useful in the production of glass for X-ray tubes and in X-ray dosimetry.

As regards the direct technical applications of the phenomena discussed here, it may be mentioned that some of them can be used for the purposes of analytical chemistry, and others underlie the design of electrical or electronic devices or switching elements. Methods of chemical analysis can be based on measurements of the residual resistance of metals, on paramagnetic resonance (for tracing paramagnetic ions present as impurities in a non-paramagnetic substance) and on thermal-conductivity measurements. We have encountered applications of the second kind in the maser and the cryotron. There is every reason to believe that more and more practical use will be made in future of those physical phenomena which occur, or are observable, only at low temperatures.

Summary. Just as the residual resistance of a metal depends on the scattering of electrons by lattice imperfections, the thermal conductivity of insulators at low temperature is determined by the scattering of phonons by the same imperfections. The behaviour of a substance is also determined, of course, by the way in which the phonon spectrum varies with temperature. As an example, the behaviour of bismuth telluride is discussed. After dealing with the subject of paramagnetic relaxation, the author discusses the importance of the temperature on the application of paramagnetic resonance and its significance in connection with a solid-state microwave amplifier (maser). Dielectric losses due to polarization mechanisms of very low activation energy (< 0.1 eV) and which can therefore only be studied at low temperatures, are found in such substances as impure quartz and non-stoichiometric or impure oxides and semiconductors.

¹³⁾ Y. Haven, Lattice imperfections in crystals, studied on alkali halides, Philips tech. Rev. **20**, 69-79, 1958/59.

ABSTRACTS OF RECENT SCIENTIFIC PUBLICATIONS BY THE STAFF OF N.V. PHILIPS' GLOEILAMPENFABRIEKEN

Reprints of these papers not marked with an asterisk * can be obtained free of charge upon application to Philips' Electrical Ltd., Century House, Shaftesbury Avenue, London W.C. 2, where a limited number of reprints are available for distribution.

- 2793:** J. G. Bonenkamp and W. Hondius Boldingh: Quality and choice of Potter-Bucky grids (Acta radiol. **51**, 479-489, 1959, No. 6; **52**, 149-157, 1959, No. 2; **52**, 241-253, 1959, No. 3).

In the first section of this paper a new and less ambiguous criterion for the quality of Potter-Bucky grids is suggested. A graphical method is given whereby the best contrast improvement can be obtained. In section II of the paper, measurements are reported on 13 grids, revealing that their contrast-improving capacity is determined mainly by their lead content. The relation between contrast, free-air dose and filters is discussed, especially at high voltages. The final section discusses the absorption of primary and scattered radiation and the relation between the number of lines per cm, ratio and lead content of grids under optimum conditions. Rules are thereby arrived at for a suitable choice of grid for a given field of application.

- 2794:** J. D. Fast and H. A. C. M. Bruning: Entkohlung und Entstickung von Eisen-Silicium-Legierungen (Z. Elektrochemie **63**, 765-772, 1959, No. 7). (Decarburization and denitriding of iron-silicon alloys; in German.)

The denitriding and decarburization of Fe-Si alloys with Si contents up to 3% by weight were investigated by heating samples for two hours in pure and in wet hydrogen at 900 °C. The rate of denitriding of Si-free iron is independent of the water content of the hydrogen. Denitriding of iron containing Si takes place in very dry hydrogen but is retarded considerably even by traces of water vapour, probably by formation of a SiO₂ skin on the surface. The rate of decarburization in very dry hydrogen is practically zero for Si-free iron but increases with increasing Si content.

It is concluded that the activation energy for the formation of CH₄ molecules on the surface is much reduced by the presence of Si. Damp hydrogen has a decarburizing effect on Si-free iron because the formation of CO on the surface requires a smaller activation energy than the formation of CH₄. Both CO and CH₄ are formed on Si-iron in damp hydrogen; decarburization would therefore be very rapid, were it not that the same retarding reaction takes place as with denitriding, viz. the

formation of SiO₂ on the surface. The SiO₂ skin practically stops the formation of CH₄ and slows down the formation of CO. The sealing-off of the surface is, however, less complete than in the case of denitriding, owing to penetration of C into the skin with the formation of SiO and CO.

- 2795:** J. L. Meijering: Störungslinien und Störungsbänder in innerlich oxydierten Kupfer- und Silberlegierungen (Z. Elektrochemie **63**, 824-829, 1959, No. 7). (Perturbation lines and bands in internally oxidized copper and silver alloys; in German.)

Perturbation of the reaction balance, e.g. of $\text{Be} + \text{O} \rightarrow \text{BeO}$, at the subscale boundary gives rise to local variations in the BeO concentration. The perturbations may be brought about by changes in gas atmosphere or in temperature, and also by a special geometry of the specimen.

- 2796:** P. Massini: Synthesis of 3-amino-1,2,4-triazolyl alanine from 3-amino-1,2,4-triazole in plants (Biochim. biophys. Acta **36**, 548-549, 1959, No. 2).

The herbicide 3-amino-1,2,4-triazole is metabolized by plants. One of the transformation products has been isolated from bean plants treated with aminotriazole, and its structure has been partially elucidated.

- 2797:** M. Avinor: Effect of aluminium on the green emission of cadmium sulphide (Physica **25**, 1095-1096, 1959, No. 11).

Note on the so-called Ewles-Kröger emission in CdS, excited by about 2.4 eV at low temperatures. It is found that activation by aluminium actually enhances the intensity of the green emission while at the same time destroying the fine structure of the emission band. There is also an enhanced emission on the verge of the infrared.

- 2798:** K. van Duuren, W. K. Hofker and J. Hermen: Compact low-level counting arrangement (Proc. 2nd United Nations int. Conf. on the peaceful uses of atomic energy, Geneva 1-13 Sept. 1958, Vol. 14, pp. 339-344; Pergamon Press, London 1959).

See Philips tech. Rev. **20**, 170-172, 1958/59.

- 2799:** W. L. Wanmaker, W. P. de Graaf and H. L. Spier: Luminescence of Pb- and Pb-Mn-activated lanthanum silicates (*Physica* **25**, 1125-1130, 1959, No. 11).

Lanthanum silicates of the composition $1\text{La}_2\text{O}_3 \cdot 1\text{SiO}_2$ and activated with Pb give under 2537 Å excitation an U.V. emission (peak wavelength 3150 Å) and activated with Pb and Mn an orange emission (peak wavelength 5950 Å). The preparation of some lanthanum silicates is described and X-ray diagrams are given (viz. of $2\text{La}_2\text{O}_3 \cdot \text{SiO}_2$, $\text{La}_2\text{O}_3 \cdot \text{SiO}_2$ and $\text{La}_2\text{O}_3 \cdot 2\text{SiO}_2$).

- 2800:** G. A. Ovezall-Klaasen and J. Halberstadt: The preparation of S^{35} - and/or Cl^{36} -labelled SO_2Cl_2 (*Int. J. appl. Radiation and Isotopes* **7**, 145-147, 1959, No. 2).

Radioactive sulphuryl chloride labelled with ^{35}S and/or ^{36}Cl can be prepared in every quantity and with every possible specific activity by mixing calculated quantities of radioactive $^{35}\text{SO}_2$ with a small excess of Cl_2 gas, or $^{36}\text{Cl}_2$ gas with a small excess of SO_2 . Mixing is done in a reaction vessel in which a small quantity of charcoal acts as a catalyst. The yields are practically 100 per cent.

- 2801:** F. L. H. M. Stumpers and R. Schutte: Stereophonische Übertragung von Rundfunksendungen mit FM-modulierten Signalen und AM-moduliertem Hilfsträger (*Elektron. Rdsch.* **13**, 445-446, 1959, No. 12). (Stereophonic transmission of radio broadcasts with frequency-modulated signals and an amplitude-modulated auxiliary carrier; in German.)

Description of a system for transmitting stereophonic signals in the FM band. With $A(t)$ and $B(t)$ as the microphone signals, the frequency modulation of the main carrier is given by $A + B + a(1 + A - B) \cos \omega t$, where a is the amplitude of the auxiliary carrier and ω its angular frequency. The equipment of the transmitter is discussed, and it is also shown that only slight modifications are required to make FM receivers, provided with two sound channels, suitable for the system discussed (this is an attractive feature of the system). A discussion is also devoted to the FM spectrum, and the expected signal-to-noise ratio is calculated.

- 2802:** J. Meltzer: Unspecific resistance mechanisms in the house-fly, *Musca domestica* L. (*Indian J. Malariology* **12**, 579-588, 1958, No. 4).

By selecting insects with a certain insecticide (i.e. treating one generation with the insecticide,

breeding the survivors, and so on for each generation), strains can be obtained that are resistant to the selecting agent. In selecting house-flies with DDT and lindane (both chlorinated hydrocarbons) the author has produced strains that show a high level of resistance to these agents. Resistance to "Diazinon" was of a lower degree, and no or only slight resistance was developed to S 17 (N,N-dimethylphenylcarbaminat). It was surprising, however, that all selected strains also showed resistance to the compounds with which they had not previously been in contact. All four were highly resistant to DDT and lindane (and also to various other chlorinated hydrocarbons used in the investigation); all four showed the same fairly low degree of resistance to "Diazinon" and scarcely any resistance to S 17. The relative toxicities of the various insecticides for the selected strains differed entirely from those for the original strain. These results suggest that this multiresistance is not so much due to specific decomposition mechanisms but rather to a generally-active mechanism. Such a mechanism might consist in a decrease of the permeability of cell membranes.

- 2803:** W. J. Oosterkamp: The concept of absorbed dose and its measurement (Symposium on quantities, units and measuring methods of ionizing radiation, Rome, April 1958, pp. 86-99; publisher Hoepli, Milan 1959).

The "International Commission on Radiological Units and Measurements" introduced in 1953 the concept "absorbed dose", which is the ratio of the energy imparted to the tissue by ionizing particles to the mass in a small volume of irradiated tissue around the point P where the dose is to be ascertained. The "exposure dose", measured in röntgens, is given by the ionization produced in air by the emission of electrons associated with the X-rays in an imaginary volume of air around the point P . The relation between these quantities is discussed and illustrated graphically for the case of a water phantom irradiated with X-rays of 0.2, 2 and 30 MeV.

- 2804:** B. Combée and K. Reinsma: Methods of measuring the effectiveness of protection against ionizing radiation (as **2803**; pp. 320-337).

Review of the requirements to be met by instruments for measuring ionizing radiation from X-ray equipment and nuclear plant, and for determining the effectiveness of installations designed to provide protection against ionizing radiation. The advan-

tages and disadvantages of existing instruments are considered, and some examples are dealt with in detail.

2805: N. V. Franssen: Enkele onderzoekingen omtrent richtingswaarneming (T. Ned. Radiogenootschap **24**, 321-335, 1959, No. 6). (Some investigations into directional hearing; in Dutch.)

A survey is given of the theory of binaural and stereophonic hearing. On the basis of an electrical model of the binaural hearing mechanism, the phenomena occurring in stereophonic reproduction are explained. Finally some methods of compressing the stereo-information are discussed.

R 399: H. J. Heijn: Representations of switching functions and their application to computers (Philips Res. Repts. **15**, 305-341, 1960, No. 4).

This thesis (Delft, 1960), continued in **R 407**, deals with switching problems in electronic computers. Of the three main elements of such computers, viz. the memory, the arithmetical unit and the control unit, the memory often consists of a matrix of square-loop ferrite rings (see e.g. Philips tech. Rev. **20**, 193, 1958/59). It is known that the arithmetical and control units could also be designed using ferrite rings in place of the usual valve or transistor flip-flops. This would involve some loss of speed, but the computer would gain in reliability. Boolean algebra, used in the design of computer circuits to determine the optimum circuit for a given operation, has to be specially adapted for application to systems using square-loop ferrite elements. The first half of this thesis is devoted to the development of an algebra based on that of Boole but adapted to systems using magnetic-ring elements. In the second half of the thesis, this algebra is used to design various magnetic-ring elements such as an adder, a counter and a decoder. Special attention is paid to the calculation of the time necessary for the transfer of carries in a binary adder. It is found that the adding time can be approximately halved when the adder is sub-divided into segments of suitable length.

R 400: S. Duinker: Durable high-resolution ferrite transducer heads employing bonding glass spacers (Philips Res. Repts. **15**, 342-367, 1960, No. 4).

The magnetic and mechanical properties of a very dense, homogeneous and fine-grained ferrite are discussed with reference to its application as a material for making transducer heads for magnetic recording and reproduction, for which it was

specially developed as a substitute for laminated metallic alloys. The technique of producing transducer gaps by preparing ultra-thin (i.e., from less than one micron upwards), well-bonding and stress-free glass layers between polished ferrite surfaces is described, and the excellent qualities of such gaps as regards their optically and magnetically measured lengths and their wear-resistance are discussed. Several novel single- and multiple-track head constructions are outlined which, compared to existing assembly methods, are characterized by reduced production costs. This is because the expensive mechanical operations (polishing and gap preparation) can be confined to the most vital parts of the head (i.e., the frontal part containing the gap) and can be done for a large number of heads simultaneously in a prefabrication stage.

R 401: G. Diemer and J. G. van Santen: Power amplifiers based on electro-optical effects; a survey (Philips Res. Repts. **15**, 368-389, 1960, No. 4).

Electro-optical effects (such as electroluminescence, photoconduction and combinations thereof) and devices based on these effects are discussed from the point of view of power amplification. Special features arise owing to the transformation of the signal (from electric to radiative form, and vice versa) and owing to the quantized nature of radiative energy transfer. A survey of applications along these lines is given.

R 402: A. J. W. Duijvestijn and B. P. A. Boonstra: Numerical evaluation of functions occurring in a study of domain configuration in thin layers of $\text{BaFe}_{12}\text{O}_{19}$ (Philips Res. Repts. **15**, 390-393, 1960, No. 4).

Two functions occurring in the theory of domain configuration in thin layers of $\text{BaFe}_{12}\text{O}_{19}$ are evaluated numerically and presented graphically.

R 403: P. B. Braun and W. Kwestroo: On some calcium-iron-oxygen compounds (Philips Res. Repts. **15**, 394-397, 1960, No. 4).

Three new calcium oxide-iron oxide compounds are stabilized by addition of small amounts of a third component:

- 1) $\text{Ca}_4\text{Fe}_{14}\text{O}_{25}$, hexagonal ($R\bar{3}c$) with $a = 6.0 \text{ \AA}$ and $c = 95.0 \text{ \AA}$. Stabilized by e.g. Y^{3+} . Ferrimagnetic, preferential plane of magnetization.
- 2) $\text{Ca}_4\text{Fe}_{14}\text{O}_{25}$, hexagonal ($P\bar{3}c$) with $a = 6.0 \text{ \AA}$ and $c = 31.6 \text{ \AA}$. Stabilized by e.g. Mg^{2+} .
- 3) $\text{Ca}_4\text{Fe}_2^{2+}\text{Fe}_{18}^{3+}\text{O}_{33}$, hexagonal ($R\bar{3}c$) with $a = 6.0 \text{ \AA}$ and $c = 62.3 \text{ \AA}$, also stabilized by e.g. Mg^{2+} ions. Ferrimagnetic, preferential plane of magnetization. Magnetic properties and X-ray data are given.